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# Polycyclic aromatic hydrocarbons associated with road deposited solid and their ecological risk: Implications for road stormwater reuse



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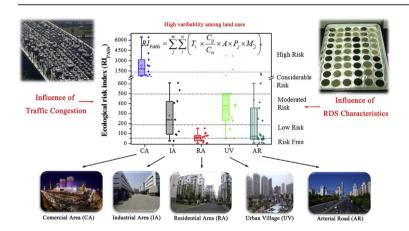
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#### HIGHLIGHTS

# GRAPHICAL ABSTRACT

- PAHs build-up on road surfaces varies with traffic and land use conditions.
- · RDS characteristics and PAH composition were considered in ecological risk assessment
- ΣPAH concentration attached to RDS cannot represent their overall ecological risk.
- Higher percentage of 5-6 rings PAHs can pose higher ecological risk.
- · TC exerts more important influences on 5-6 rings PAHs build-up compared with TV.



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### ABSTRACT

Reusing stormwater is becoming popular worldwide. However, urban road stormwater commonly contains toxic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), which could undermine reuse safety. This study investigated pollution level of PAHs and their composition build-up on urban roads in a typical megacity in South China. The potential ecological risk posed by PAHs associated with road deposited solid (RDS) was also assessed. Results showed that ecological risk levels varied based on different land use types, which could be significantly influenced by the composition of PAHs and characteristics of RDS. A higher percentage of high-ring PAHs, such as more than four rings, could pose higher ecological risk and are more likely to undermine stormwater reuse safety. Additionally, the degree of traffic congestion rather than traffic volume was found to exert a more significant influence on the generation of high-ring PAH generation. Therefore, stormwater from more congested roads might need proper treatment (particularly for removing high-ring PAHs) before reuse or could be suitable for purposes requiring low-water-quality. The findings of this study are expected to contribute to adequate stormwater reuse strategy development and to enhance the safety of urban road stormwater reuse.

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## 1. Introduction

As an underutilized resource, stormwater has received significant attention for water reuse (Liu et al., 2015). This attention is particularly important in stormwater management and safeguarding urban water quality (Shannak et al., 2014). However, unsatisfactory stormwater quality can constrain its reuse because of the presence of pollutants that originate from urban surfaces, particularly roads (Li et al., 2007; Mannina, 2005). Road stormwater runoff can be recycled if these pollutants can be properly removed (Field and Fan, 1981). In this situation, reusing road stormwater entails precise gauging of the amount of pollutants found on the surface of roads. Thus, a thorough understanding of the pollutants build-up processes is necessary because of stormwater wash-off that could affect the concentration of undesirable elements. Moreover, since these pollutants could potentially pose ecological risks, which vary with total pollutant toxicity, loads and mobility (Zhao et al., 2014), their ecological risk should be also taken into consideration to ensure stormwater reuse safety.

Previous research identified road deposited solids (RDS), which consist of vehicle exhaust, tire wear, soil dust, and erosion of infrastructures, as primary pollutant contributors to road stormwater runoff (Gunawardana et al., 2012; Miguntanna et al., 2010; Thorpe and Harrison, 2008). A variety of toxic pollutants has been identified as specific pollutants of road runoff because of their close relationship with RDS (Jiang et al., 2014; Wang et al., 2011; Zheng et al., 2014). Among these pollutants, polycyclic aromatic hydrocarbons (PAHs) are among the persistent organic pollutant (POP) candidates that have become topics of interest because of their ecological risk potential (Vanderzalm et al., 2011). Sixteen parent PAHs that have different properties, such as benzene ring numbers (Di Toro et al., 2000), have been identified by the United States Environmental Protection Agency as priority pollutants. Previous researchers have noted that the number of benzene rings plays an important role in influencing the toxicity of PAHs (Delistraty, 1997). Boström et al. (2002) claim that PAHs with more than four rings contain a "bay-region" and a "K-region," which account for metabolic formation of bay- and K-region epoxides that perform covalent binding with DNA. Therefore, high-ring PAHs possess higher toxic potential compared with those with low-ring numbers (Nisbet and LaGoy, 1992; Samanta et al., 2002). This observation highlights that the composition of PAH species (such as different benzene ring number) on road surfaces could result in differences in their ecological risk even though the total PAH loads (the sum of 16 species) might be the same.

The study of risk assessment encourages significant ideas on safeguarding strategies for water environment (Caeiro et al., 2005; Wang et al., 2011); thus, precise evaluation of ecological risk of PAHs is needed for effective stormwater reuse implementation (Liu et al., 2015). Previous studies have focused on the risk assessment of PAHs attached to the incremental lifetime cancer risk of RDS to human beings rather than their possible ecological risk posed to the water environment resulting from the wash-off process of PAHs (Jiang et al., 2014; Lorenzi et al., 2011). Moreover, ecological risk assessment, such as geo-accumulation index, enrichment factor, and Nemerow synthetic pollution index, focus on pollutants such as heavy metals (Cheng et al., 2007; Muller, 1969; Shi et al., 2010; Zhao et al., 2014), and only consider pollutant concentrations or toxicology hazards. Thus, evaluating the ecological risk of PAHs associated with RDS has not received attention. This research gap motivates the development of a new approach to evaluating the ecological risk of PAHs associated with RDS, which comprises PAH composition, toxicity, and RDS characteristics.

Since the road site characteristics could influence PAH load and composition (Boonyatumanond et al., 2007; Lee and Dong, 2010), thereby affecting the levels of PAH concentrations in stormwater. An important consideration is the proper determination of potential ecological risk in reusing road stormwater generated from various road sites. For example, Wang et al. (2011) reported that industrial areas were responsible for producing high loads of PAHs on road surfaces because of largescale industrial release, while RDS obtained from heavy traffic areas and busy commercial streets indicated substantial high-ring PAH loads (Dong and Lee, 2009). These outcomes prove the influence of traffic characteristics, such as volume, congestion, prescribed speed limit, and age of vehicles (Goonetilleke et al., 2009; Gunawardena et al., 2014) as well as land use on PAH load and composition. Therefore, these factors influence the ecological risk posed by PAHs associated with RDS. In this context, identifying key factors that influence potential ecological risk posed by PAHs associated with RDS is essential in developing stormwater reuse strategies for various road sites.

This paper presents the outcomes of an extensive study on the buildup of PAHs on road surfaces within a range of land uses in a typical megacity in South China. The primary objectives are (1) to characterize PAH pollution level and composition on different land uses, (2) to assess potential ecological risk posed by PAH species, and (3) to identify the key factors that influence the potential ecological risk posed by PAHs. The new finding is expected to help enrich the application of stormwater reuse strategy and to ensure reuse safety.

## 2. Materials and methods

#### 2.1. Site selection

The research study was conducted in South China, which is a subtropical zone with abundant annual rainfall (ranging from 1400– 2000 mm) but usually suffers from water scarcity. Shenzhen, which is a typical megacity and highly developed in South China (the extent of about 2000 km<sup>2</sup> and a population of over 11 million), was selected as the study site. In order to investigate PAHs build-up, 64 urban road surfaces selected across Shenzhen City encompassed a combination of Commercial Area (CA), Industrial Area (IA), Residential Area (RA), Urban Village Area (UV) and Arterial Road Area (AR). Land use types and traffic characteristics were selected as potential influential factors of PAHs build-up. Fig. 1 shows these study sites while Table S1 in the Supplementary information presents the detailed information of the selected study roads and their characteristics.

#### 2.2. Sample collection

RDS were sampled using a dry and wet vacuuming system to ensure the collection efficiency of both coarse and fine particles (Herngren et al., 2006). This sampling approach was found to have an over 90% efficiency in collecting and retaining RDS with typical particle size distribution (Mahbub et al., 2010). Since the composition of RDS can vary across the road surface (Deletic and Orr 2005), the plot which has an area of  $2 \times 1.5$  m<sup>2</sup> and encloses with aluminum alloy frame was placed at the middle of each road surface to maintain the consistency of the build-up sampling. As noted by previous research studies (Egodawatta et al., 2013; Liu et al., 2016), pollutant build-up on road surfaces approaches a constant value with the increase in antecedent dry days and after about 12 to 14 dry days it asymptotes to an almost constant value. Therefore, RDS sample collection was conducted on an antecedent dry period of 14 days. Accordingly, a total of 64 build-up samples were collected from the 64 selected road surfaces.

Given that traffic characteristics could significantly influence the PAHs build-up processes on urban road surfaces, traffic parameters were incorporated into the analysis. The traffic parameters included traffic volume (TV) and traffic congestion coefficient (TC). TV could be used to determine the total exhaust volume (Lee and Dong, 2010) while TC represents the vehicle driving patterns such as go-stop frequency (Gunawardena et al., 2014). Both TV and TC were measured at a certain time (16:00 to 17:00) on each study site. TV was collected by manual counting while TC was obtained from the Transport

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