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Comparison of pollutants in stormwater runoff from asphalt and concrete roads

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Abstract. Controlling non-point source pollutants (NPSPs) is critical in achieving good surface water quality; the contribution of road runoff has recently received increased attention. This study monitored the runoff characteristics of NPSPs, including suspended solids, particle size distribution, heavy metals, organic matter, and nutrients, from asphalt and concrete roads. Water quality parameters, including biochemical oxygen demand (BOD), chemical oxygen demand (COD), and nutrients of the receiving reservoir, were also investigated. During the first flush, the changes in pollutant concentrations over time were higher on concrete roads than asphalt roads. Concentrations peaked over a short period, while an increased pollutant concentration may be present several days after rain. The runoff concentration and particle size distribution were higher on concrete roads, whereas the concentrations of heavy metals were similar in asphalt and concrete roads. The organic matter concentration of asphalt roads was higher, or identical, to that in the first flush from concrete roads; this may be associated with the road location. Water quality analysis of the reservoir showed relatively good results for BOD, COD, and nutrient concentrations. Road construction was a factor that determined the characteristics of NPSPs in road runoff.

Keywords: asphalt; concrete; non-point source pollutant; road runoff

1. Introduction

Rapid industrial growth and a growing population have substantially altered land use, environmental quality, and increased natural resource consumption (Müller *et al*. 2020). Land use modifications are associated with urbanization, leading to changes in aquatic ecosystems, water quality deterioration, and increased flooding from higher stormwater production (Zgheib *et al*. 2012). Ongoing urbanization has increased the area of impervious surfaces, including buildings, roads, and parking areas. Various non-point source pollutants (NPSPs), including organic matter, nutrients, pathogens, heavy metals, pesticides, oil, and grease, accumulate in the dry season. These NPSPs are then transported during rainfall, increasing peak runoff pollutant concentrations (Björklund *et al*. 2018, C. Kim *et al*. 2014). These pollutants directly impact ecosystem integrity, the stability of surface water and ultimately impair watersheds (Park and Park 2015, Son *et al*. 2015, Walsh *et al*. 2012, Maestre and Pitt, 2006, Mitchell 2005). Nutrients are one of the main offending pollutants, as they are major contributors to biodiversity loss in aquatic ecosystems and eutrophication; the latter is regarded one of the greatest threats to water systems (Wu *et al*. 2012, Bricker *et al*. 2008). Additionally, increased rainfall intensity due to climate change causes periods of extreme flow, short-duration runoff, and higher peak runoff pollutant concentrations due to the rapid transport of highly concentrated NPSPs that have been accumulating on urban surfaces (Kim *et al*. 2017, Semadeni-Davies *et al*. 2008).

Point source pollutant is where a known volume of the contaminant is discharged from a single identifiable source. By contrast, NPSPs are an aggregate of small contaminant inputs distributed throughout a watershed (Leon *et al*. 2001). As these pollutants are transported during rainfall, they are difficult to predict and quantify, largely due to the extreme changes in daily and seasonal weather conditions (Poudel *et al*. 2013). As a result, past efforts to reduce pollutant loads entering watersheds have mainly focused on point source pollutants and have failed to adequately address the impact of NPSPs. It is necessary to manage NPSPs with the same level of focus and supporting research (Niraula *et al*. 2013, Jang and Kang 2009).

The literature review contains numerous studies on rainwater and stormwater runoff from highways. The runoff from roofs and lawns on the research center campus and heavily trafficked road in Beijing, China, demonstrates the importance of the first flush in urban stormwater treatment (Yufen *et al*. 2008). A study on variations in highway stormwater runoff and treatment performance concerning porous friction courses was conducted at four sites in Auckland, New Zealand (Moores *et al*. 2013). Li *et al*. (2019) investigated the influence of various design conditions on urban runoff and NPSPs. In South Korea, NPSPs accounted for approximately 68% of the pollution load in the river in 2010, and were expected to account for approximately 72% in 2020 due to the expansion of impervious surfaces in development projects (Kim and Lee, 2018, Park *et al*. 2013, S. 2010). Previous studies have analyzed runoff contaminated withs NPSPs from urban land

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use in South Korea (Rhee *et al*. 2012). Various measures and projects to reduce NPSPs have been implemented; however, their effectiveness has been questioned. This is because it is difficult to monitor and control the effects of NPSPs as they are influenced by the meteorological, geological, hydrological, demographics, type of watershed, and land use of the watershed (Kang *et al*., 2014). The biochemical oxygen demand (BOD) of four major rivers in South Korea has improved due to government-led efforts to improve water quality. However, improvements to the chemical oxygen demand (COD), including nonbio-degradable polluting materials, has deteriorated or become stagnant since the 2000s. To ensure effectiveness of pollutant reduction measures and minimize uncertainty, there is a need to actively conduct research on various aspects of NPSP management, along with periodic policy evaluations.

Roads are paved with impervious materials, including asphalt and concrete. Highly impervious surfaces significantly increase runoff volume by preventing soil infiltration and creating high velocity flows that easily transport solids or scour contaminants from surfaces; this occurs almost immediately following the beginning of a rainfall event (Ma *et al*. 2002, Sleavin *et al*. 2000). In general, pollutant concentrations from urban highway runoff exceed those in non-urban highway runoff. In particular, contaminants from the road that accumulate on a bridge may discharge directly into rivers and lakes; this can significantly impact nearby water systems (Swadener *et al*. 2014, Kayhanian *et al*. 2003). Accordingly, the Ministry of Environment in Korea has installed NPSP treatment facilities, including water source protection areas, specific countermeasure areas, and riparian buffer zones; these are located up and downstream of intake facilities. (15 km upstream and 1 km downstream) (WEPA 2015, Jeon *et al*. 2019). Moreover, a study carried out a trend analysis of stormwater runoff quality (Salim *et al*. 2019), finding that optimization of the stormwater piping network affected the quality of surface water bodies (Kim *et al*. 2019). Understanding stormwater runoff is an essential factor in evaluating long-term water quality management, as investigated by Jung *et al*. (2019).

The concentrations and loads of road NPSPs in runoff are affected by various spatio-temporal factors, including antecedent dry days, traffic volume, land use, rainfall events, road maintenance, and gradient (Yoon *et al*. 2010). Generally, dust mass on road surfaces and the strength of the first flush increases with the duration of the dry period prior to rainfall events. Thus, the mass of pollutants washed off during a storm event is dependent on the number of antecedent dry days (Lee and Lee 2009, Kim *et al*. 2006). There are also high concentrations of pollutants on roads in urban areas with heavy traffic volumes (Lee *et al*. 2007). Areas with a considerable amount of grass, such as parks, emit fewer pollutants than other urban areas. By contrast, commercial and industrial areas have a higher mass of NPSPs in the stormwater runoff because they emit more air pollutants (Shin *et al*. 2004). Furthermore, differences in runoff may occur despite similar rainfall intensitiesbecause additional factors affect runoff; this includes rainfall duration, intensity, and slope steepness (Shin *et al*. 2010). Despite ongoing research concerning these different factors, there are few studies on the transport of NPSPs from paved roads. This study aims to compare and analyze the characteristics of NPSPs in runoff from asphalt and concrete roads that discharge into water systems during rainfall events. This understanding will help design of an efficient management plan for NPSPs.

2. Materials and methods

2.1 Study site location

To examine the water quality and pollutants in runoff from paved roads, paved asphalt (site A) and concrete (site B) areas were selected based on access and safety. As these sites were adjacent, they were similar in terms of many conditions, including the number of antecedent dry days, traffic volume, land use, road maintenance, and rainfall event frequency, intensity and duration. Table 1 lists the characteristics of each site.

Site A was located near the mountain, and site B was a bridge road that passes through the reservoir, as shown in Fig. 1. At both sites, pollutants that had accumulated on the road surface were discharged directly into the water system during rainfall. The survey site, where runoff was transported from the road surface to the reservoir during rain, was located on the national highway 34, 1277-2, Songcheon-dong, Andong-si, Gyeongsangbuk-do. The average daily traffic volume on this road was 7378 vehicles, which is relatively low. The average annual rainfall was 876 mm, the maximum altitude of the measuring section was 119 m, and the minimum height was 99 m; this latter parameter is representative of a relatively gentle slope. According to the Road Drainage Facilities Design and Management Guide from the Ministry of Land, Infrastructure and Transport, the runoff coefficient ranges of asphalt and concrete are 0.70–0.95 and 0.80–0.95, respectively (Lee 2013).

2.2 Sampling

Sampling was conducted at each site for 3 h until 5 mm

Table 1 Characteristics of sample sites

Site	Pavement type	Area (m^2)	Number of lanes	Runoff coefficient
A	Asphalt	8400	2 (One way)	0.8
B	Concrete	7000	2 (One way)	0.9
	Number of antecedent dry days	Rainfall (mm)	Duration (h)	Average rainfall intensity (mm/h)
A & B	13	42.0	16	2.6

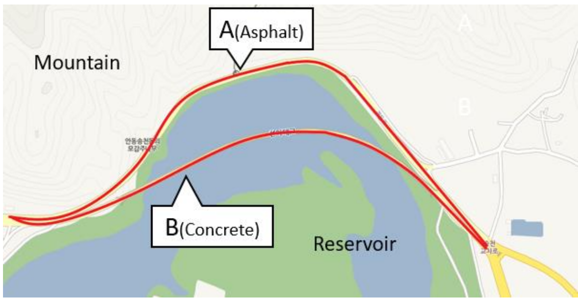

Fig. 1 Mapping of the testing site

of rainfall was collected. The first sampling was conducted immediately after rain as the first flush contains a high concentration of pollutants. Subsequently, eight samples were collected following 5, 10, 20, 30, 60, 120, and 180 min of rainfall. Samples were then immediately stored in an icebox, transported, and analyzed. In addition to measuring the particle size distribution at the same site, an artificial rainfall experiment was conducted with components similar to actual rainwater.

2.3 Analytical methods

The pollutants were analyzed for the following: suspended solids (SS), particle size distribution, BOD, UV absorbance at 254 nm (UV_{254}), total nitrogen (TN), total phosphorus (TP), pH, alkalinity, and heavy metals (Fe, Ni, Cu, and Zn). This analysis was carried out to compare the runoff characteristics of NPSPs from asphalt and concrete roads. The Korean standard methods for examining water and wastewater were used to analyze BOD, SS, TN, TP, and alkalinity. Particle size distribution was analyzed using a particle size analyzer (PSA Mastersizer 2000, Malvern Instruments, UK), while UV_{254} was measured using a spectrophotometer (UV-VIS Spectrophotometer, HACH, USA). The pH was measured with a pH meter (S.G./Orion Star A325, Thermo Fisher Scientific, USA), and heavy metals were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS 7500, Agilent, USA) (Korea Ministry of Environment, 2018).

2.4 Event mean concentration

Generally, the arithmetic mean concentration is a reliable measure when the time interval between sampling is constant. However, it is not applicable for rainfall runoff because there are changes in runoff volume and pollutant concentrations over time; as such, the sampling interval is not consistent. The event mean concentration (EMC) is a valuable parameter for predicting the pollutant load (Maniquiz *et al*. 2010). The EMC was calculated to understand the effects of stormwater runoff to water systems as a function of the concentration and volume of BOD, SS, TN, and TP. As constant monitoring was not carried out in this study, it was assumed that the outflow at a specified time was the same (Ahn *et al*. 2013), and may be calculated using the following equation:

$$EMC = \sum_{t=0}^{t=T} C_t \,.\, Q_t \Big/ \sum_{t=0}^{t=T} Q_t \qquad (1)$$

where EMC represents the event mean concentration (mg/L); and C_t and Q_t denote the pollutant concentration (mg/L) and runoff volume (L) at specific time intervals, respectively.

3. Results and discussion

3.1 Rainfall characteristics

The rainfall characteristics at the time of sampling are shown in Table 1, while Table 2 compares the initial analyzed rainwater characteristics with those of previous studies. Sample pH ranged from 4.1 to 7.4, electrical conductivity (EC) ranged from 3 to 68 µS/cm, SS was 0.8 mg/L, turbidity ranged from 0.42 to 3.88 NTU, and BOD was 1.3 mg/L. These ranges were caused by the variations in the spatio-temporal and rainfall event characteristics, including the number of antecedent dry days, rainfall amount, and duration. These were essential factors to consider when analyzing the characteristics of NPSPs.

3.2 Comparative analysis of particulate matter in runoff

Water quality characteristics, including SS, BOD, UV_{254}, TN, TP, pH, alkalinity, heavy metals, and the particle size distribution of road surface runoff based on pavement construction, were compared for each site. A first flush occurred at each site, and the peak concentration of soluble solids at site B (67.0 mg/L) was higher than that of site A (30.5 mg/L) (Fig. 2). The concentration changed

Table 2 Comparison of the initial rainwater qualities with previous studies

Parameters	Current study	Previous studies				Range
		(An *et al.* 2016)	(Lee *et al.* 1999)	(Lee *et al.* 2004)	(Kim *et al.* 2014b)	
pH	6.0	5.8	4.3–5.0	4.76	6.8–7.4	4.3–7.4
Alkalinity (mg/L)	8.7	-	-	-	-	8.7
EC (µs/cm)	5.5	10–50	23–68	11.42	-	5.5–68
UVA @254 nm	0.002	-	-	-	-	0.002
SS (mg/L)	0.8	-	-	-	-	0.8
Turbidity (NTU)	-	0.75	-	-	0.42–0.64	0.42–0.75
BOD (mg/L)	1.3	-	-	-	-	1.3

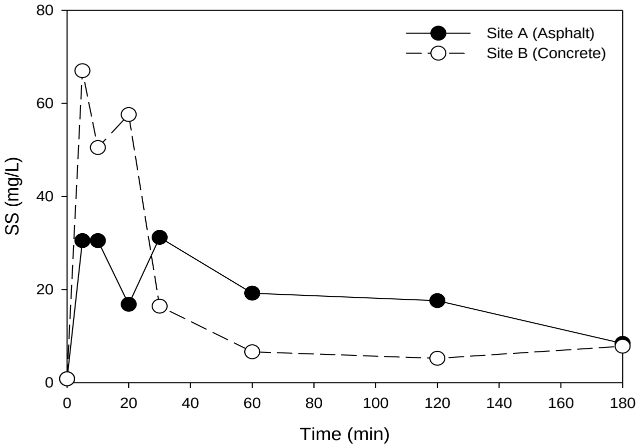

Fig. 2 Soluble solids concentration in runoff at both sampling sites

Table 3 Comparison of particle size distributions during the first flush

Site	D_{10} (µm)	D_{50} (µm)
A (Asphalt)	10.7	31.6
B (Concrete)	11.2	43.6

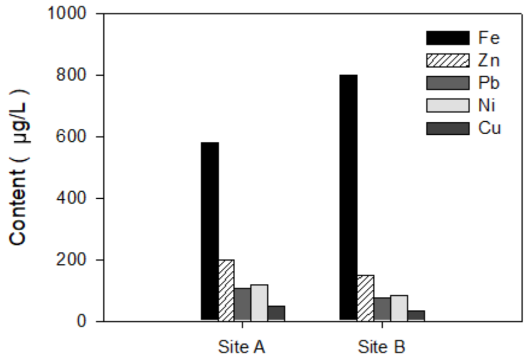

Fig. 3 Event mean concentrations of heavy metals in runoff at both sampling sites

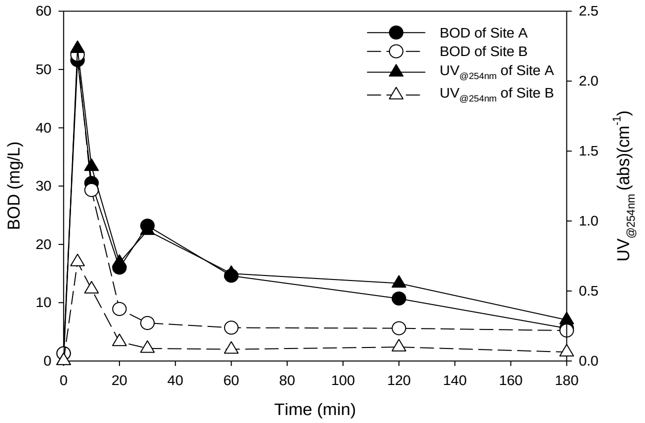

Fig. 4 Changes in BOD concentration and UV absorbance in runoff at both sites

overtime at site B (from 5.2 to 67.0 mg/L), and was also larger than site A, ranging from 8.4 to 30.5 mg/L.

The pollutant concentration at site B was lower than that at site A after 30 min of rainfall. The EMC at sites A and B were 18.81 and 22.54 mg/L, respectively; thus, there was a higher pollutant concentration in the runoff from site B. More impurities accumulated due to the relatively high roughness coefficient and were rapidly transported from the uniform road surface during rainfall. Moreover, various NPSPs, including nutrients, heavy metals, and hydrocarbons, were absorbed into particulate matter and released during rain. During the process of runoff production and movement, pollutants and particulate matter undergo adsorption, desorption, deposition, and resuspension, where the particle size distribution has a decisive effect (Zhang *et al.* 2017). As such, we analyzed the particle size distribution of samples collected during the rainfall experiment (Table 3).

The particle size distributions at site A were 10.7, 31.6, and 387 µm for D10, D50, and D90, respectively; at site B, these distributions were 11.2, 43.6, and 507 µm, respectively. The diameter of particulate matter at site B was relatively significant compared to that at site A. Heavy metals, including Fe, Zn, Pb, Ni, and Cu, were analyzed to identify the characteristics of runoff as it relates to heavy metals in paved road materials (Fig. 3). The Fe content at site B (799 µg/L) was prominent compared to site A (581 µg/L), while the remainder were all higher at site A: Zn was 201 µg/L at site A and 151 µg/L at site B, Pb was 109 µg/L at site A and 77 µg/L at site B, Ni was 118 µg/L at site A, and 83 µg/L at site B, and Cu was 48 µg/L at site A and 36 µg/L at site B. Heavy metals on road surfaces originate from various sources, including tire wear, oil and grease, the corrosion of vehicle bodies, steel structures, external inflow, and heavy metals that have been adsorbed into eroded soil that is transported to the road from the mountain. These metals were discharged during rainfall and resulted in a higher concentration of heavy metals at site A.

3.3 Comparative analysis of organic matters, nutrients, and ionic materials in runoff

The peak runoff concentration of BOD was 52.5 mg/L at site B, and 51.6 mg/L at site A (Fig. 4); the change in concentration overtime at site B was more pronounced. The EMC was higher at site A (15.09 mg/L) than at site B (9.8 mg/L). Similarly, the peak UV_{254}, indicative of organic matter derived from natural ecosystems, at sites A and B, was 2.232 and 0.708 abs, respectively. The EMC at site A (0.675 mg/L) was significantly higher than that at site B (0.148 mg/L).

The TN and TP exhibited similar changes in concentration overtime compared to other pollutants (Fig. 5). The EMCs of TN and TP at site A were 1.65 and 0.28 mg/L, while those at site B were 1.03 and 0.21 mg/L, respectively; thus the concentrations of both pollutants were higher at site A. Previous comparisons of NPSP characteristics in runoff from asphalt and concrete demonstrate that most pollutants on a concrete road have higher EMCs. By contrast, this study observed higher concentrations on the asphalt road for organic matter and nutrients as per the literature (C. Kim *et al.* 2014). This may

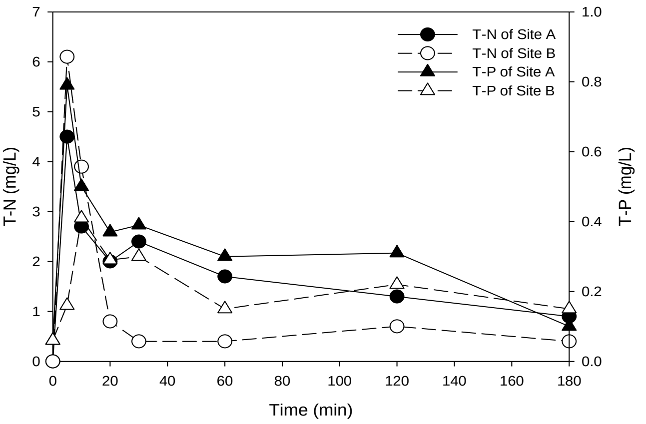

Fig. 5 Changes in TN and TP concentrations in runoff at both sampling sites

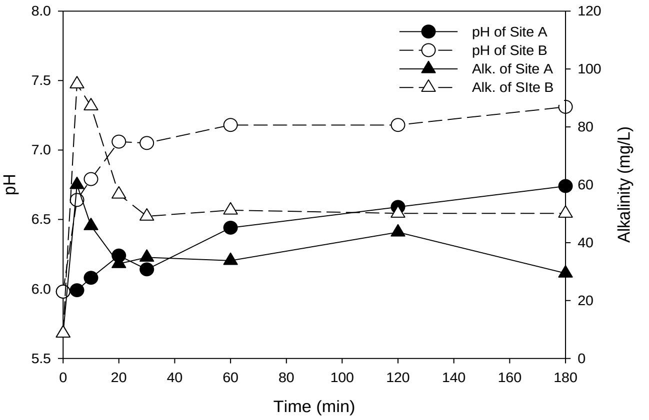

Fig. 6 Changes in pH and alkalinity of runoff at both sampling sites

be due to organic matter, and nutrients being transported onto site A from nearby mountainous areas. As site B is a road built through the reservoir, the potential for tis transport is relatively low. This supports the argument stated in McKnight *et al.* (1997) that other organic matter is introduced from sources such as nearby forests, resulting in higher concentrations.

The pH and alkalinity represent ionic materials, where the former tended to increase at both sites; in contrast to other parameters, which tended to decrease over time (Fig. 6). Event mean concentrations were 6.46 at site A and 7.15 at site B. Alkalinity exhibited similar characteristics, with a higher initial runoff concentration at site B; there was a substantial difference in the EMCs between the sites, being 35.89 mg/L at site A and 53.94 mg/L at site B. The reaction between carbon dioxide in the atmosphere and the hydration products in concrete results in the production of calcium carbonate. This reduces the alkalinity inside the concrete; as such, pH and alkalinity increase because the alkali component on the road surface dissolves in stormwater during rainfall, affecting the present study (Cavalline 2018).

4. Conclusions

This study investigated the NPSPs transported from adjacent asphalt and concrete roads with similar traffic volumes. A number of conclusions were drawn after analyzing and comparing the pollutant characteristics in the runoff from both roads. During the first flush, the change in pollutant concentration overtime was more significant on the concrete road than its asphalt counterpart because of its flatter surface; this resulted in the more rapid transport of NPSPs. The SS and particle size distribution of particulate matter were higher on concrete roads because of the increased friction between these roads and vehicle tires. However, the heavy metal concentrations were similar at both sites. The trend in the organic matter and nutrient concentrations indicated that the asphalt road had a higher, or similar, first flush concentration compared to the concrete road; however, these pollutants were more rapidly transported over time. This is likely due to the transport of organic matter and nutrients from the mountainous areas near the asphalt road. Water quality analysis showed relatively good results for BOD, COD, TN, and TP. This may be due to the relatively low traffic density at the targeted sites and differences in the various rainfall events and topographic characteristics. It is recommended that continuous monitoring of the NPSP characteristics for runoff in the area is carried out over a long period of time to design an appropriate approach to treat road runoff.

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Author contributions

S. Kim (Ph.D. candidate) and J. Lee (BS student) conducted experimental work, organized data, and wrote the first draft of the manuscript; M. Yaqub (Postdoc researcher), undertook data analysis, rewriting, revisions, and updated the manuscript writing; W. Lee (Professor) revised and finalized the manuscript.

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