AES BIOFLUX
Advances in Environmental Sciences - International Journal of the Bioflux Society

Analysis of road-deposited sediments for heavy metal pollutants in bridge sidewalks of Iloilo City, Philippines

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Abstract. Road-deposited sediment is one of the major sources of heavy metals pollution of urban waterways. This study determined the presence of heavy metals: Cr, Cu, Pb, and Zn in road-deposited sediments collected from bridge sidewalks in Iloilo City, Philippines, using cold HCl as extractant to represent the "leachable" form of the metals, which is considered the most bioavailable. In general, mean heavy metal concentrations in sediments from all bridge sites were significantly different from background values. Concentrations of Cu and Pb were found to be up to 7 and 6 folds higher than the background values, respectively. Zn had the highest concentration and up to 14 fold higher than the background. A significant correlation was observed between Cr and Zn, and Cu and Pb indicating similar industrial and traffic sources. High positive correlations (R^2 = 0.650–0.915) were observed between heavy metals concentrations and the number of vehicles per hour passing through the bridges. With the exception of Pb, the road-deposited sediment samples collected in Iloilo City showed much lower heavy metal concentrations compared to those reported for urban environments in other countries. Over-all, all heavy metals concentrations were observed to be higher for all sites during the dry season. The metals analyzed in this study have all been detected in the Iloilo River estuary sediment and water at concentrations exceeding the guideline levels of the Philippine Department of Environment and Natural Resources (DENR).
Key Words: heavy metals, road-deposited sediments, Iloilo City, Philippines, urban dust, pollution.

Introduction. The contamination of urban areas with heavy metals presents a significant hazard in the normal functioning of urban ecosystems (Taylor & Owens 2009; Davis et al 2001). Road-deposited sediment is one of the major sources of heavy metals pollution of urban waterways (Yisa 2010; Herngren et al 2006). Heavy metal contaminated sediments accumulated on road surfaces and curbs can be easily mobilized and transported through urban storm water run-off, to surface water bodies, affecting benthic community structures and functions of the receiving water bodies (Zhu et al 2008; Duzgoren-Aydin et al 2006; Sutherland & Tolosa 2000). Accumulation in fish and other food sources in the receiving water may pose potential public health risks associated with metal toxicity (Wei & Yang 2010; Sutherland & Tolosa 2000). Thus, it is important to minimize pollutant inputs from land-based sources to aquatic ecosystems (Forman & Alexander 1998).

Metals in road-deposited sediments in urban areas may come from different sources, including abrasion of vehicular components and their exhaust emissions, tire and road surface wear (Yisa 2010; Zhang & Wang 2009; Adachi & Tainosho 2004; Councell et al 2004; Lehner et al 1999). Motorized vehicles have become the foremost mode of transportation. The consistent rise in the number of motored vehicles leads to the increase of traffic-related pollutant emission in the environment, especially in urban areas adjacent to traffic activities (Hidayah et al 2008; Zhu et al 2008).

A number of studies on heavy metal contamination of road-deposited sediments in urban environments have been carried out in recent years (Wei & Yang 2010; Yisa 2010; Zhang & Wang 2009; Hidayah et al 2008; Zhu et al 2008; Duzgoren-Aydin et al 2006;

Herngren et al 2006; Sutherland & Tolosa 2000). However, most of these studies were conducted in developed countries, with very different urban environments from developing countries, particularly in tropical environments (Duzgoren-Aydin et al 2006; Jaradat & Momani 1999). The contamination of road-deposited sediments and street dusts receives very little attention in the Philippines, where vehicular heavy metal emission is expected to be high, mainly due to the proliferation of junk shops, car repair shops and more importantly vehicles, particularly jeepneys (the main mode of transportation in the country), which are 20 years old or older. As far as we know, this is the first report on heavy metal contamination in road-deposited sediments in the Philippines.

Located 470 km from Manila on the Island of Panay, Iloilo City is the residential, financial, commercial, governance and educational hub for the Western Visayas region. Iloilo City consists about 180 barangays and six districts with an estimated population of 424,619 in 2010, an area of 56.1 sq km, and an annual growth rate of 1.49 % from 2000 to 2010 (NSCB-PSA 2013). Due to its population growth and industrialization, more roads and infrastructures such as bridges are being built in the already congested city. Iloilo River is the main surface water body in the city, stretching a distance of 11 km from the mouth at port of Iloilo to the Municipality of Oton where it joins another river system (the Batiano River). Like most urban waterways, it is a sink to many toxic chemicals.

This study determined the presence of heavy metals such as Cr, Cu, Pb, and Zn in road-deposited sediments collected from bridge sidewalks at Iloilo City. Specifically, it determined the concentration of heavy metals in samples collected from four bridges in Iloilo City (Forbes Bridge, Iloilo Bridge, Jalandoni Bridge, Quirino-Lopez Bridge) using cold HCl as extractant to represent the "leachable" form of the metals, which is considered the most bioavailable (Sutherland et al 2004). Temporal (wet and dry seasons) variation of heavy metal concentrations in road-deposited sediments was also determined. The metals levels in road-deposited sediments were also correlated to the average number of vehicles passing through the bridges. As far as we know, this is the first report on heavy metal contamination in road-deposited sediments in the Philippines.

Material and Method

Study site. Samples were collected from 4 sites, consisting of 2 high traffic density bridges and 2 low traffic density bridges located at the city of Iloilo. These include the Iloilo Bridge (I) and Forbes Bridge (F) for high traffic volume, and Jalandoni Bridge (J) and Quirino-Lopez Bridge (QL) for low traffic volume. The list of bridge sampling sites with their corresponding bridge names, location, Global Positioning System (GPS) coordinates, and descriptions are shown in Table 1. Soil dust samples were also collected from a control site, an open field intended for housing project, which is located in an area far from industrial factories and road traffic. Heavy metals levels determined in this site were considered background levels.

Table 1

Bridge sampling sites with their corresponding descriptions

Sampling site	*Location (see Figure 1)*	*GPS coordinates*	*Description*
Iloilo Bridge	I	10°42'8.81"N , 122°33'12.54"E	High traffic volume, ~>2500 vehicles/hr
Jalandoni Bridge	J	10°42'6.72"N , 122°33'39.67"E	Low traffic volume, ~<1000 vehicles/hr
Forbes Bridge	F	10°42'22.59"N , 122°34'2.94"E	High traffic volume, ~>2500 vehicles/hr
Quirino-Lopez Bridge	QL	10°42'6.70"N , 122°34'16.05"E	Low traffic volume, ~>1000 vehicles/hr

Sample collection and processing. Each bridge was divided equally into 3 sections at both sides making up 6 sections. From each section, street dust samples were collected using plastic brush and plastic dustpan along the surface of the sidewalk and transferred into clean polyethylene bags for transport to the laboratory. A sample composite for each bridge was made by mixing all the samples taken from the 6 sections of the bridge. The composite samples were oven-dried for at least 24 hours. After drying, the samples were sieved through a 180µm mesh size nylon sieve, and grinded using mortar and pestle for homogenization prior to chemical analysis. The first sampling period was on the month of October 2009 and the second was on the month of January 2010. Sampling was done on a dry sunny weather or at least a few days after the last rainfall.

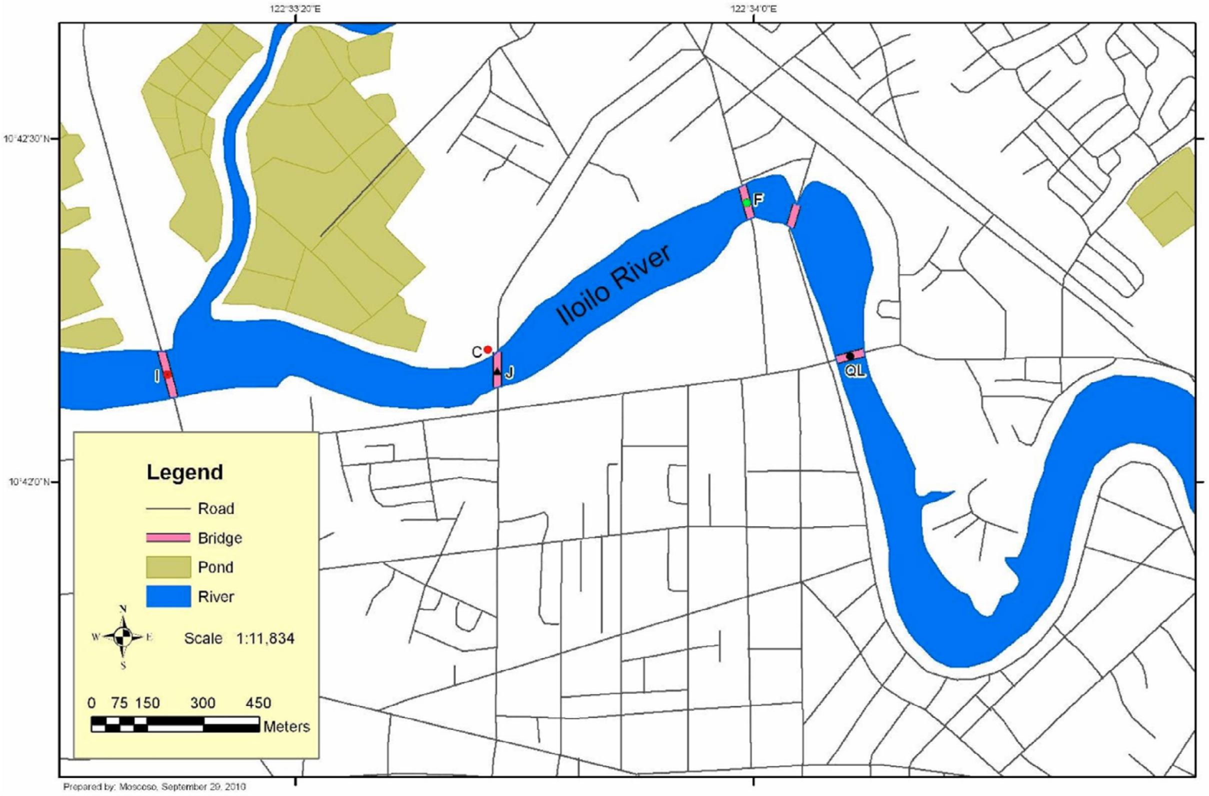

Figure 1. Map of Iloilo City, Philippines with sampling sites (prepared by Moscoso 2010).

Chemical analysis. The digestion method was adapted from Sutherland et al (2004) with slight modifications. Briefly, approximately 1.0 g of sediment sample was added into 20 mL of 0.5 M HCl in 25 mL screw capped test tubes. The samples were then shaken for 1 hour at room temperature. The shaking mechanism was done with the use of an improvised shaker setup. After digestion, the mixtures were filtered with Whatman 41 filter paper into 50 mL volumetric flasks and diluted to the mark with deionized water. The samples were then transferred into polyethylene bottles and stored prior for FAAS analysis. Each sample composite consists of three replicates. All glasswares and containers were thoroughly cleaned and acid washed before using.

The heavy metals (Cr, Cu, Pb, Zn) in the sediment samples were analyzed using an Flame Atomic Absorption Spectroscopy (FAAS) (Varian SpectrAA 55B FAAS, Australia) using the working conditions listed in Table 2. Standard solutions for Cr, Cu, Pb, and Zn were prepared by a series of dilutions of each of their respective stock solutions (1,000 mg L^{-1}). Instrument detection limit (IDL) for Cr, Cu, Pb and Zn are 4.35, 0.128, 0.742 and 0.585 µg L^{-1}, respectively.

Table 2
Flame Atomic Absorption Spectrophotometer (FAAS) working conditions with respect to each heavy metal analyzed

Working condition	*Cr*	*Cu*	*Pb*	*Zn*
Lamp current (mA)	7	4	5	5
Fuel	Acetylene	Acetylene	Acetylene	Acetylene
Support	Air	Air	Air	Air
Flame stoich	Reducing	Oxidizing	Oxidizing	Oxidizing
Wavelength (nm)	357.9	324.7	217.0	213.9
Slit width (nm)	0.2	0.5	1.0	1.0
Optimum working range (µg mL^{-1})	0.06-15	0.03-10	0.1-30	0.01-2

Data analysis. The mean and standard deviation (SD) of the heavy metal concentrations were determined through summary statistics using Microsoft Office Excel 2007. A comparison was made using One-way Analysis of Variance (ANOVA) to determine the significant difference between the concentrations of heavy metals present in the dust samples of the four bridges. T-test was also used to determine if there was a significant difference in the temporal concentrations of heavy metals in dust samples on each bridge. Statistical analysis was performed using SPSS v16.0 Software for Windows™.

Results and Discussion. This study determined the presence of heavy metals: Cr, Cu, Pb, and Zn in road-deposited sediments collected from bridge sidewalks in Iloilo City, Philippines using cold HCl as extractant to represent the "leachable" form of the metals, which is considered the most bioavailable (Sutherland et al 2004).

The mean heavy metal concentrations in road-deposited sediment collected along bridge sidewalks from the four bridges during the two sampling periods are given in Table 3. In general, heavy metal mean concentrations in sediments from all bridge sites are significantly different from the background values except for Cr in samples collected from site J. Concentrations of Cu and Pb were found to be up to 7 and 6 folds higher than the background values, respectively. Zn had the highest concentration and up to 14 fold higher than the background. A previous study, reported similar observations in urban run-off sediments with Zn found to have highest concentration and the most widely distributed among metals (Jartun et al 2008).

Table 3
Mean heavy metal concentrations (mg kg^{-1}) ± SD in road-deposited sediment samples collected from Iloilo Bridge (I); Jalandoni Bridge (J); Forbes Bridege (F); Quirino-Lopez Bridge (QL) and a Control site (C) during wet (October 2009) and dry season (January 2010).

Sampling site	*Cr*	*Cu*	*Pb*	*Zn*
C	3.31±0.34	5.44±0.93	9.24±1.28	9.55±1.43
		Wet season		
I	3.80±0.08	18.31±5.39	30.51±3.51	97.87±2.57*
J	3.05±0.15*	20.54±4.60	33.67±4.13	64.53±4.21*
F	4.06±0.24	27.46±4.86	44.26±2.57	111.77±4.14*
QL	4.31±0.35	61.57±12.85*	132.91±2.44*	137.08±1.34*
		Dry season		
I	4.14±0.01	19.32±8.95	39.30±3.07*	129.31±3.52*
J	3.21±0.24*	14.39±1.09	15.82±1.83*	68.43±7.95*
F	5.81±0.34*	45.64±3.85*	61.33±2.02*	176.51±0.48*
QL	4.69±0.07	62.47±4.48*	115.00±3.18*	154.27±3.14*

* indicates significant difference ($p < 0.05$) in levels of the metal between sampling sites for a given sampling period.

Temporal (wet and dry seasons) variation of heavy metal concentrations in road-deposited sediments was also determined. The mean heavy metal concentrations in road-deposited sediments showed similar trend for the two sampling periods (Table 3). However, heavy metal concentrations were found to be significantly higher in road-deposited sediments collected during the dry season (Table 3).

The road-deposited sediments collected from site J during the wet season had significantly ($p < 0.05$) lower Cr concentration compared to those from other sites (Table 3). Similarly, Cr concentrations in road-deposited sediment samples were lowest and highest in sites J and F, respectively, for samples collected during the dry season. The lower Cr concentration could be attributed to the low traffic volume in site J compared to the other sites (Table 4). These observations suggest that Cr in road-deposited sediments likely originated from traffic sources (Yisa 2010; Zhang & Wang 2009; Hidayah et al 2008; Zhu et al 2008; Lehner et al 1999). Studies reported that air conditioning coolants, engine parts and brake emissions are among the most common anthropogenic sources of Cr in road-deposited sediments (Yisa 2010; Zhang & Wang 2009; Lehner et al 1999). In addition, the type of vehicles passing on the bridge could be a significant contributing factor in this observation.

Table 4

Pearson's correlation matrix for the heavy metal concentrations and number of vehicles

Sampling Site	*Cr*	*Cu*	*Pb*	*Zn*
Cr				
Cu	0.698			
Pb	0.554	0.960*		
Zn	0.958*	0.769	0.700	
No. of vehicles	0.595	0.087	-0.144	0.569
No. of vehicles[a]	0.797	0.650	0.892	0.915

* correlation is significant at the 0.05 level (2-tailed); [a] QL is excluded in the analysis.

The road-deposited sediment samples collected from site QL showed significantly ($p < 0.05$) higher Cu and Pb concentrations compared to the other three bridge sites in both sampling times. Concentrations of Cu in road-deposited sediment samples from F and QL were found to be significantly higher in the dry season samples. Road-deposited sediment samples collected from all four bridges have Zn concentrations significantly different from one another (Table 3).

The levels of Cu and Pb in road deposited sediments could be attributed to vehicle emissions such as break wear and leaded gasoline, respectively (Yisa 2010; Zhang & Wang 2009; Jartun et al 2008; Lehner et al 1999). While the difference in Zn concentrations could be attributed to the tire wear and corrosion of galvanized parts of vehicles since vehicle tires are directly in contact with the bridge or the road itself which would likely be deposited in dusts (Kim et al 1998). Shinggu et al (2007) attributed high concentrations of Zn, Pb, Cu, and Cr in street dust samples collected from the streets of Mubi, Adamawa State, Nigeria, to exhaust emissions from vehicles, oil spillage of gasoline, diesel, engine oils and lubricating oils, brake emissions, bearing wear and rusting of non-coated auto body parts.

When compared to levels reported for urban environments in other countries, the mean concentrations of heavy metals in road-deposited sediments for all bridge sites in Iloilo City showed much lower heavy metal concentrations except for Pb, which was higher than the levels reported in two cities in China (Leung et al 2008; Wei et al 2010; Herngren et al 2006). This indicates that Iloilo City is probably still using more leaded gasoline compared to Urumqi and Xuzhou cities in China. However, it should be noted that different metals extraction techniques will give you different results, and the technique used in this study is cold HCl extraction to represent only the "leachable" form of the metals, which is considered the most bioavailable (Sutherland et al 2004). Nevertheless, comparing the values found in this study with those reported in other cities gives us a general idea on the extent of heavy metal contamination in various urban environments.

The correlations between heavy metal concentrations and the number of vehicles per hour in all sites are shown in Table 4. Results show that there is a significant correlation ($p < 0.05$) between Cu and Pb, indicating that these heavy metals might share a common source such as traffic emissions. Possible sources of these heavy metals could be attributed to older (>15 years old) public utility jeepneys (PUJs) due to corrosion of vehicular body parts and the use of leaded fuels. Moreover, Cr and Zn are also significantly correlated ($p < 0.05$). Levels of these heavy metals may be attributed mainly to industrial activities and other anthropogenic sources in the area (Kim et al 1998). Sources of Cr could be from taxis and private vehicles that use air coolants. In general, there is no significant correlation between the number of vehicles and heavy metal concentrations (Table 4). However, a significant correlation is observed if the QL site is excluded from the analysis. That is because in spite of its low traffic volume, high metal concentrations were observed in the road-deposited sediments from this site (Tables 3 and 4). It should be noted that heavy truck loaders and tankers pass through this bridge, which could have contributed to the high concentrations of heavy metals.

Conclusions. This study determined the concentration of heavy metals (Cr, Cu, Pb, and Zn) in road-deposited sediments deposited along bridges in Iloilo City. Metal enrichment was around 1, 7, 6, and 14 folds higher than the background levels for Cr (3.31 mg kg^{-1}), Cu (5.44 mg kg^{-1}), Pb (9.24 mg kg^{-1}), and Zn (9.55 mg kg^{-1}), respectively. These metals are easily leachable once they enter the surface water bodies (e.g. Iloilo river), where they may become bioavailable to aquatic organisms and subsequently to humans. The metals analyzed in this study have all been detected in the Iloilo River estuary sediment and water at concentrations exceeding the guideline levels of the Philippine Department of Environment and Natural Resources (DENR).

All heavy metals concentrations were observed to be higher for all sites during the dry season. A significant correlation was observed between Cr and Zn, and Cu and Pb indicating possible industrial and traffic sources. High positive correlations (R^2 = 0.650–0.915) were also observed between heavy metals concentrations and the number of vehicles per hour passing through the bridges.

The contamination of road-deposited sediments and street dusts receives very little attention in the Philippines. This is the first report that we know of on heavy metal contamination in road-deposited sediments in the Philippines. Future studies should include more metals and frequent monitoring of road-deposited sediments. In addition, road-deposited sediments should also be further divided based on particulate matter sizes.

Acknowledgement. The authors would like to thank Mr. Allan Dino Moscoso for the preparation of site map.

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Received: 20 January 2014. Acepted: 01 February 2014. Published online: 21 February 2014.
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How to cite this article:
Taberna Jr. H. S., Nillos M. G. G., Gamarcha L. T., Pahila I. G., Gastar A. N., 2014 Analysis of road-deposited sediments for heavy metal pollutants in bridge sidewalks of Iloilo City, Philippines. AES Bioflux 6(1):69-75.