Faculty of Resource Science and Technology

DETERMINATION OF HEAVY METALS IN WATER AND SEDIMENT OF BATANG BALEH

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Determination of Heavy Metals in Water and Sediment of Batang Baleh

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LIST OF ABBREVIATIONS

AAS	Atomic Absorption Spectrophotometer
Cd	Cadmium
CF	Contamination Factor
CRM	Certified Reference Material
Cu	Copper
Fe	Iron
HCl	Hydrochloric Acid
HEP	Hydroelectric Power
Hg	Mercury
Hg Analyzer	Mercury Analyzer
HNO_3	Nitric Acid
I_{geo}	Geo-accumulation Index
Mn	Manganese
Mo	Molybdenum
NWQS	National Water Quality Standards
Ni	Nickel
Pb	Lead
PLI	Pollution Load Index
SCORE	Sarawak Corridor of Renewable Energy
ST	Station
Zn	Zinc

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Determination of Heavy Metals in Water and Sediment of Batang Baleh

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ABSTRACT

This study determines the concentration of selected heavy metals (Cd, Cu, Fe, Pb, Mn, Ni, Zn, and Hg) in water and sediment samples in order to assess the heavy metal pollution status in Batang Baleh. Composites of water and sediment samples were collected simultaneously at 18 sampling stations. Selected metals were analyzed using an atomic absorption spectrophotometer and a mercury analyzer. Metal concentrations of water ranged between 0.039-0.435 mg/L for Fe, 0.005-0.026 mg/L for Pb, 0.062-0.097 mg/L for Mn, 0.007-0.008 mg/L for Ni, and 0.006-0.015 µg/L for Hg. Meanwhile, for sediment, it ranged between 11.58-20.75 mg/kg for Cu, 12626.70-38623.37 mg/kg for Fe, 2.26-18.90 mg/kg for Pb, 35.51-389.77 mg/kg for Mn, 7.43-36.11 mg/kg for Ni, 17.78-58.48 mg/kg for Zn, and 0.0005-0.0747 mg/kg for Hg. The average metal concentrations in the water were lower than the National Water Quality Standards, Canadian Water Quality Guidelines, and National Recommended Water Quality Criteria by USEPA. The average concentration of studied metals in sediment was well below the Ontario Sediment Standards and Canadian Freshwater Sediment Guidelines. Assessment of contamination status by contamination factor, geo-accumulation index, and pollution load index suggests that there is no risk of contamination at the moment.

Key words: heavy metals, river, water, sediment, Batang Baleh

ABSTRAK

Kajian ini menunjukkan kepekatan logam berat terpilih (Cd, Cu, Fe, Pb, Mn, Ni, Zn, dan Hg) dalam sampel air dan sedimen untuk menilai status pencemaran logam berat di Batang Baleh. Komposit sampel air dan sedimen telah dikumpulkan secara serentak di 18 stesen. Logam terpilih telah dianalisis dengan menggunakan AAS dan penganalisis Hg. Kepekatan logam air adalah antara 0.039-0.435 mg/L untuk Fe, 0.005-0.026 mg/L untuk Pb, 0.062-0.097 mg/L untuk Mn, 0.007-0.008 mg/L untuk Ni, dan 0.006-0.015 µg/L untuk Hg. Sementara itu, bagi sedimen, ia adalah antara 11.58-20.75 mg/kg bagi Cu, 12626.70-38623.37 mg/kg bagi Fe, 2.26-18.90 mg/kg bagi Pb, 35.51-389.77 mg/kg bagi Mn, 7.43-36.11 mg/kg untuk Ni, 17.78-58.48 mg/kg bagi Zn, dan 0.0005-0.0747 mg/kg bagi Hg. Secara keseluruhan, kandungan logam berat di dalam air adalah lebih rendah daripada Piawaian National bagi Kualiti Air, Garis Panduan Kualiti Air Kanada, dan Kriteria bagi Kualiti Air yang dicadangkan oleh USEPA. Kepekatan purata logam berat dikaji dalam sedimen adalah jauh di bawah Piawaian Sedimen Ontario dan Garis Panduan Sedimen Air Tawar Kanada. Penilaian status pencemaran oleh nilai-nilai CF, I_{geo} dan PLI menunjukkan bahawa tidak ada risiko pencemaran buat sementara waktu ini.

Kata kunci: *logam berat, sungai, air, sedimen, Batang Baleh*

1.0 INTRODUCTION

Heavy metals, also known as trace elements, exist naturally in the ecosystem from processes such as rock weathering which leads to the geo-chemical recycling of heavy metals (Muwanga, 1997; Zvinowanda *et al.*, 2009). However, rapid development and industrialization drives the surge in anthropogenic sources of heavy metals being washed off into the aquatic environment. Heavy metals possess substantial environmental threats due to its acute and chronic toxicity, bioaccumulative nature, widespread occurrence, non-biodegradable, and non-thermo-degradable characteristics (Karishma & Prasad, 2014; Yu *et al.*, 2008). Furthermore, heavy metals are generally distributed among the aqueous phase and the sediments during their transport in the aquatic environment. Besides the running river water that carries mobilized heavy metals, sediments serve as the source and medium to trap heavy metals. Heavy metals are usually incorporated into sediments *via* processes such as ion exchange, adsorption, complexation, mineral precipitation. These elements then became available for uptake by aquatic organisms and ends up in human through the progression of food chain. Heavy metals could result in severe health effects when consumed excessively. For instance, cadmium exposure might cause kidney damage, lung cancer, and hearing dysfunction (Järup, 2003).

Various studies have been conducted to assess the degree of heavy metal contamination in the river systems. Most of the riverine studies are concerned with the total metal concentration by investigating metal contents in water, sediment, and/or aquatic biota. Examples of researches conducted in Malaysia include spatial variability of metals in surface water and sediment in the Langat River and geochemical factors that influence the water-sediment interactions (Lim *et al.*, 2012), concentration of selected heavy metals in water of the Juru River (Idriss & Ahmad, 2013), and bioaccumulation of selected metals in freshwater Haruan fish collected from the Pahang River basin (Jalal *et* *al.*, 2013). Most of these studies were dedicated on the total metal content in sediments, aquatic organisms, and freshwater bodies for a better interpretation on the heavy metals' ecotoxicological potential.

With reference to the Sarawak Corridor of Renewable Energy agenda, a series of hydroelectric power (HEP) projects has been planned. One of the scheduled hydroelectric dams is located at Batang Baleh, about 95 km from its confluence with the Rajang River in Kapit Division (Sarawak Energy Berhad, 2013). Batang Baleh has remains largely undisturbed for decades despite the logging activities (World Wildlife Fund (WWF), 2012). However, as the development of HEP embarks, heavy metals contamination could become a concern (Aprile & Bouvy, 2008; Jain *et al.*, 2012; Maitera *et al.*, 2011).

To date, no study has been done to investigate the status of heavy metals in Batang Baleh. Therefore it is essential to capture the baseline data before the development of hydroelectric dam. This would serve as background information for future monitoring.

The main objective of this study is to determine the distribution of heavy metals in the river water and sediment of Batang Baleh.

2.0 LITERATURE REVIEW

2.1 Heavy Metals

In general, there are two categories of contaminants that can be found in the river water, sediment, and biota: organic and inorganic contaminants (Miller & Miller, 2007). One of the inorganic contaminants that affect the river systems considerably are heavy metals. Heavy metals are defined as high density elements that are typically belong to the transition group of periodic table (de Vries *et al.*, 2002). According to Sardar *et al.* (2013), heavy metals are trace elements with a density of at least five times than that of water. There are three general categories of heavy metals in the aquatic environment: metals in suspended particulates (> 0.45 ppm), metals adsorbed onto hydrous oxides and humic colloids, and metals found in the sediment and in the true solution.

Heavy metals possess significant environmental threats due to its toxicity, wide variety of sources, non-biodegradable characteristics, and accumulative behaviors (Jula, 1971; Yu *et al.*, 2008). Sources of heavy metals in the environment include mining and effluents from industrial activities such as smelting, oil refining, petrochemical manufacturing and chemical manufacturing, untreated sewage sludge, agricultural wastewater, as well as scatter sources such as metal piping, traffic and by-products from combustion activities in coal burning power plants (Dinis & Fiúza, 2011). Despite the toxicological effects of heavy metals, elements such as manganese (Mn), copper (Cu) and molybdenum (Mo) play a vital role in physiological functions of living cells and regulation of many biochemical mechanisms when present in trace amount (Hall, 2002; Singh *et al.*, 2011). Table 1 (Manahan, 2000) shows the sources, effect and significance of heavy metals found as contaminants in natural waters.

Table 1: Trace elements found as contaminants in natural waters (Manahan, 2000).

Element	Sources	Effect and significance
Arsenic	Mining byproduct; chemical waste	Toxic; possibly carcinogen
Beryllium	Coal; industrial waste	Toxic
Boron	Coal; detergents; wastes	Toxic
Chromium	Metal plating	Essential as Cr (III); toxic as Cr (IV)
Copper	Metal plating; mining; industrial waste	Essential trace element; toxic to plants and algae at higher levels
Iron	Industrial waste; mining; fuels	Essential nutrient; damage fixtures by staining
Lead	Industrial wastes; mining; fuels	Toxic; harmful to wildlife
Manganese	Industrial wastes; acid mine water; microbial action	Toxic to plants; damages fixtures by staining
Mercury	Industrial waste; mining; coal	Toxic; mobilized as methyl compounds by anaerobic bacteria
Molybdenum	Industrial wastes; natural source	Essential to plants; toxic to animals
Selenium	Natural sources; coal	Essential at lower levels; toxic at higher levels
Zinc	Industrial waste; metal plating; mining; plumbing	Essential element; toxic to plants at higher levels

The foremost threats to human health from heavy metals are related to exposure to arsenic, cadmium, selenium, lead and mercury (Dinis & Fiúza, 2011; Järup, 2003; Prasher, 2009). Other heavy metals that are of great concern to human health and to the environment include copper, manganese, nickel, iron, and zinc.

2.1.1 Arsenic

Arsenic is one of the most widely dispersed metalloid. It is found naturally occurring in rock, soil, water and air. Apart from existing naturally in the environment, arsenic is released in larger amounts into the environment *via* occasions such as volcanic activity, rocks erosion, forest fires, and human activity (Martin & Griswold, 2009). Inorganic arsenic which is odorless and tasteless is present in groundwater in several countries (Järup, 2003; National Institute of Environmental Health Sciences (NIEHS), 2014). It is acutely toxic and carcinogenic. The presence of arsenic in water supplies might lead to exposure in shellfish, cod, and haddock in the form of organic arsenic (Prasher, 2009). Inorganic arsenic may be transformed to more mobile and lethal methylated organic derivatives such as methylarsenic acid and dimethylarsenic acid (Subramanian, n.d.). In short, consumption of arsenic-polluted seafood and drinking water from groundwater are major pathways of arsenic exposure in humans. According to Subramanian (n.d.), the first symptom of arsenic exposure is discoloration of the skin. It later develops into skin cancer and eventually affects the lungs, liver, kidneys and bladder.

2.1.2 Cadmium

Cadmium, in its purest form, is a soft silver-white metal that is found naturally in ores together with copper, lead, and zinc (Cope *et al.*, 2004). Cadmium usually exists in combinations with other elements. For instance, cadmium oxide, cadmium chloride, and cadmium sulfide are commonly found in the environment. The industrial application of cadmium includes as a pigment in paint and plastic products, in electroplating, and in making alloys and re-chargeable nickel-cadmium batteries (Cope *et al.*, 2004; Järup, 2003). The environmental exposure to cadmium is mostly due to groundwater contamination as a result of smelting activities, various industrial applications and the usage of fertilizer from sewage sludge (Martin & Griswold, 2009). Acute health effects of cadmium exposure include kidney damage, lung cancer, and hearing dysfunction (Prasher, 2009). Long-term high cadmium exposure might lead to skeletal damage. The disease itai-itai was first reported in Japan in 1950s as a result of consumption of cadmium-contaminated rice.

2.1.3 Lead

Lead is classified as a very soft metal. Due to its long-term and widespread usage, it becomes one of the most ubiquitous lethal metals. As reported by Agency for Toxic Substances and Disease Registry, ATSDR (2007), lead is ranked the second in the ATSDR's list. Lead is widely used in various industrial applications such as as fuel additives, as pigments in paints, in batteries, and construction materials. Other sources of lead include lead pipes and glazed ceramic food container (Cope *et al.*, 2004; Subramanian, n.d.). The general population is exposed to lead *via* probable pathways such as air, water, or food sources. According to Prasher (2009), lead essentially affects the entire human body systems. The environmental exposure to chronic low levels of lead has been associated to a broad range of metabolic disorders and neuropsychological deficits (Nriagu, 1988). The health effects of lead exposure are obvious mainly in three organ systems, which is the hematological system, the central nervous system and the renal system (Hutton, 1987).

2.1.4 Mercury

Mercury occurs in the environment in three major chemical forms, namely the elemental mercury (Hg^0), inorganic mercurous (Hg^+) and mercuric (Hg^{2+}) salts, and organic methylmercury (CH_3Hg) and dimethylmercury (CH_3HgCH_3) compounds. As discussed by Basak (2009) and Järup (2003), mercury enters the environment through industrial discharges such as chloro-alkali industry which uses mercury as the electrode in the electrochemical process for manufacturing of chlorine, electrical apparatus of production plants, and from the agricultural industry which applies a large amount of fungicide for seed dressings. Cope *et al.* (2004) states that of more worrying issues from environmental contamination is the exposure to organic mercury compounds. Inorganic mercury is converted by the action of sulfate-reducing bacteria in the aquatic sediments, to form methylmercury, a highly lethal form of organic mercury which is readily absorbed across membranes (Subramanian, n.d.). Methylmercury is very stable and have a tendency to accumulate in the food chain. Humans are largely exposed to mercury through food, fish being a main source of methylmercury exposure (World Health Organization (WHO), 1990). Typically, methylmercury compound does not cause severe health risk. However, those who consume fish more often tend to have higher risk of being affected by the compound. The health effects of mercury exposure include some neurological and psychological signs such as personality change, sleep disturbance, depression, kidney failure, and paresthesia (Prasher, 2009).

2.1.5 Copper

Copper are defined as a reddish metal that can be found naturally in rock, soil, water, sediment and air at a small amount. According to ATSDR (2004), anthropogenic activities such as mining of copper and other metals, industrial effluents, waste dumps, domestic wastewater, burning of fossil fuels, wood manufacture, and production of phosphate fertilizer introduces copper into the environment. Since copper is widespread in the environment, humans are easily exposed to the element by breathing air, drinking water, food consumption, and by physical contact with soil, water and other copper-comprising substances (ATSDR, 2004). Copper is essential at lower levels but harmful at higher dosage. As discussed by Ashish *et al.* (2013), long term exposure to copper dust could cause irritation, headaches, dizziness, nausea, and diarrhea. High intakes of copper could lead to severe liver and kidney damage and eventually death.

2.1.6 Manganese

Manganese occurs naturally in rocks and soil but it does not occur as pure metal in the environment. It usually exists in combination with other elements such as oxygen, sulfur, and chlorine (ATSDR, 2012). Anthropogenic sources of manganese could cause enrichment of the element in the environment. Manganese is primarily used in manufacturing of ferromanganese steels to improve hardness, stiffness, and strength. Other applications of manganese are electrolytic manganese dioxide for use in batteries, fungicides, antiknock agents, pigments, wood preservatives, and coating welding rods (Bradl, 2005). It is an essential element at lower levels but toxic at higher levels; a high concentration of manganese could affect the respiratory tract and the brain. The general populations are exposed to manganese primarily *via* food sources. According to ATSDR (2012), the signs of manganese poisoning include hallucinations, poor memory and nerve damage. Manganese can also cause Parkinson, lung embolism and bronchitis.

2.1.7 Nickel

Nickel is found in the environment primarily combined with elements such as oxygen or sulfur to form oxides or sulfides (ATSDR, 2005). It is found in soil and is also released from volcanoes. Most of the nickel in the environment is found in soil and sediments. This is because nickel tends to attach to particles containing iron or manganese, which are common constituents of soil and sediments. Examples of industrial application of nickel are: as an alloy in the steel industry, electroplating, Ni/Cd batteries, and pigments for paints and ceramics (Bradl, 2005). ATSDR (2005) states that anthropogenic sources of nickel includes auto exhaust emission, fertilizers, superphosphate, food processing, industrial wastewater discharge, stainless steel cookware, and burning of fossil fuel. The common health effect of nickel is the allergic reaction which causes skin rash at the contact site. Overexposure of nickel could lead to symptoms such as headache, dizziness, shortness of breath, vomiting, and nausea, whereas the delayed effects are of chest pain, coughing and bluish discoloration of the skin under severe circumstances, delirium, convulsions, and even death (ATSDR, 2005).

2.1.8 Iron

Iron is the fourth most abundant element in the Earth's crust. It is found naturally in rocks and soils. According to Wright and Welbourn (2002), sediments of aquatic environment are usually comprised of high concentrations of iron. Weathering of rocks and soil erosion causes mobilization of particulate forms of iron. Besides, it also releases soluble iron that enters the surface water directly or indirectly. Examples of activities that introduce anthropogenic sources of iron into the environment are: metal smelting and refining activities, steel production, and metal plating (Bradl, 2005). Improper waste disposal practice might result in mobilization of ferrous iron under acidic and/or anaerobic conditions (Wright & Welbourn, 2002). Iron is an essential element for various metabolic functions, particularly in redox reactions. Toxicity of iron in humans is rather unusual and rare. Chronic iron toxicity is frequently linked to excessive dietary iron intake associated to large consumption of red meats or dietary iron supplements.

2.1.9 Selenium

Selenium is an element which is widely distributed in the Earth's crust and is commonly presence in the sedimentary rock (ATSDR, 2003). In the environment, much of selenium in rocks is combined with sulfide, silver, copper, lead, and/or nickel minerals. As discussed by Subramanian (n.d.), elemental selenium becomes slowly accessible to plants from soils. Selenous acid ($H_2SeO_3^-$) is relatively mobile in the aqueous environment and is available to plants. Besides, SeO_4^{2-} is a strong oxidizing agent and is reducible to SeO_3^{2-} under most environmental conditions. Selenium is used in the electronic and glass industries, inorganic pigments, rubber production, stainless steel, lubricants, and as a component of fungicides (ATSDR, 2003; Bradl, 2005). People are exposed to low levels of selenium *via* food, water, and air. Selenium is an essential nutrient for humans and animals. However, at higher concentrations, selenium becomes highly toxic. Selenium exposure at chronic level might cause selenosis, chemical pneumonia, bronchitis, lower blood pressure, and possibly carcinogenic (Subramanian, n.d.).

2.1.10 Zinc

Zinc, in its pure elemental state, appears as a bluish-white, shiny metal. It is one of the most common elements in the Earth's crust. Most zinc ore that occurs naturally in the environment is present as zinc sulfide. Zinc can be found in air, soil, water, and is even present in all foods (ATSDR, 2005). Zinc enters the environment as a result of both natural processes and anthropogenic activities. Often, human activities such as mining, electroplating (galvanization), production of alloys, dry cell batteries, white paints, ceramics, rubber, wood preservation and manufacturing and dyeing of fabrics, introduces zinc into the environment and causes metal pollution (Bradl, 2005). Inhalation of excessive zinc dust or fumes from mining or smelting could lead to a condition known as the metal fume fever. However, this symptom is reversible as soon as the exposure to zinc ceases. Other health effects that are mentioned by ATSDR (2005) are stomach cramps, nausea, vomiting, possible anemia and damages to the pancreas as well as decreasing high-density lipoprotein cholesterol levels.

2.2 Rivers

Rivers are the most important freshwater reserve for humans. According to Juanico and Agno (1987), about 75 % of Earth's surface is covered in water whereas the remaining is of different type of landscapes. Around 97 % of water on Earth's surface is saline and are therefore deemed improper for domestic, industrial and/or agricultural purposes. Of the remaining, only 2.5 % of the Earth's water resource is freshwater and approximately 0.49 % of all freshwater is found in rivers.

As development and industrialization progresses, the health of rivers systems had been altered dramatically by anthropogenic activities (Meybeck & Vörösmarty, 2005; Wang & Chen, 2009). One of the greatest concerns is the heavy metal pollution in river systems (Edokpayi *et al.*, 2014; Malik *et al.*, 2014; Rahman *et al.*, 2013). Heavy metal pollution in rivers occur generally due to atmospheric deposition, increased rate of industrial activities, discharge from municipal wastewater treatment plant, rapid urban and agricultural progression and/or non-point source runoff consisting of pesticides and fertilizers (Kaki *et al.*, 2011; Sanayei *et al.*, 2009). Most of the dissolved metals ended up in rivers and are then absorbed onto colloid particulates (van Aardt & Erdmann, 2004). Under conditions such as high alkalinity, they may be precipitated (Jennett & Foil, 1979).

Physical movement of chemical species in a river is basically by advection. This phenomenon is due to the gravitational movement of masses of water (Manahan, 2011). Advection is a process defined as the transportation of masses of solute towards downstream by the mean of flow velocity of rivers (Runkel & Bencala, n.d.). In general, advection results in quick mixing and dilution due to velocity shear in which water move at different rates. Furthermore, turbulent mixing and diffusive transport of dissolved species and colloidal particles also contributes to the mixing process as well. These processes ultimately cause the pollutants such as the heavy metals to be introduced into the river system and become wide spread as it flows towards downstream (Manahan, 2011).

Table 2 (Malaysia Department of Environment (DOE), 2010) shows the National Water Quality Standards (NWQS) for the concentration of heavy metals in rivers of Malaysia. Table 3 (DOE, 2010) further illustrates the definition of the classes.

Table 2: NWQS for metals' concentration in rivers of Malaysia (DOE, 2010).

Parameters	Unit	Classes				
		I	IIA/IIB	III#	IV	V
Al	mg/L		-	- (0.06)	0.5	
As	mg/L		0.05	0.4(0.05)	0.1	
Ba	mg/L		1	-	-	L
Cd	mg/L		0.01	0.01* (0.001)	0.01	E
Cr (IV)	mg/L	N	0.05	1.4 (0.05)	0.1	V
Cr (III)	mg/L	A	-	2.5	-	E
Cu	mg/L	T	0.02	-	0.2	L
Hardness	mg/L	U	250	-	-	S
Ca	mg/L	R	-	-	-	
Mg	mg/L	A	-	-	-	
K	mg/L	L	-	-	-	
Fe	mg/L		1	1	1 (leaf) 5 (others)	A
Pb	mg/L		0.05	0.02* (0.01)	5	B
Mn	mg/L		0.1	0.1	0.2	O
Hg	mg/L	L	0.001	0.004 (0.0001)	0.002	V
Ni	mg/L	E	0.05	0.9*	0.2	E
Se	mg/L	V	0.01	0.25 (0.04)	0.02	
Ag	mg/L	E	0.05	0.0002	-	
Sn	mg/L	L	-	0.004	-	
U	mg/L	S	-	-	-	IV
Zn	mg/L		5	0.4*	2	
B	mg/L		1	- (3.4)	0.8	
Cl	mg/L		200	-	80	
NO_3	mg/L		7	-	5	

* = At hardness 50 mg/L $CaCO_3$ # = Maximum (unbracketed) and 24-hour average (bracketed) concentrations

Table 3: NWQS class definitions (DOE, 2010).

Class	Definition
I	• Water bodies of excellent quality, most suitable for water extraction for human consumption. • Conservation of natural environment. • Water supply I – Practically no treatment necessary (except by disinfection or boiling only). • Fishery I – Very sensitive aquatic species.
IIA	• Water bodies of good quality. • Water supply II – Conventional low cost treatment required. • Fishery II – Sensitive aquatic species.
IIB	• Recreational use with body contact. • Excessive treatment, which acquires relatively high costs, is required for human consumption.
III	• Water bodies that are polluted. • Water supply III – Extensive/advanced treatment required. • Fishery III – Common of economic value, and moderately tolerant aquatic species; livestock drinking.
IV	• Water bodies that are heavily polluted. • Irrigation purposes only.
V	None of above.

2.3 Sediments

According to Manahan (2000), sediment is defined as layers of relatively finely divided matter casing the bottoms of rivers, streams, lakes, reservoirs, bays, estuaries, and oceans. In general, sediments are made up of mixtures containing fine-, medium-, and coarse-grained particles. These particles are comprised of clay, silt and sand mixed with organic matter which may vary in composition from pure mineral matter to predominantly organic matter. As discussed by the United States Environmental Protection Agency, USEPA (2014), weathering and erosion of rocks or unconsolidated deposits causes sediment to be released into the environment. It is usually suspended, transported or deposited by water.