

Spatial and Temporal Variation on Distributions of Dissolved and Particulate Arsenic and Mercury in Sungai Kuantan, Pahang

(Perbezaan Temporal dan Reruang bagi Taburan Logam Arsenik dan Merkuri Terlarut dan Berpartikel di Sungai Kuantan, Pahang)

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ABSTRACT

It is important to monitor the concentration of toxic metals in the Sungai Kuantan as it serves many communities in terms of domestic, fisheries and agriculture purpose. In order to determine the distributions of dissolved and particulate As and Hg in Sungai Kuantan and evaluate its changes temporally and spatially, water samples were collected from the surface and bottom layers in a grid of 9 stations from estuary towards the upstream of Sungai Kuantan from May 2012 till October 2012. The dissolved metals were pre-concentrated using Chelex-100 while particulate metals were digested using Teflon bomb and subsequently were analyzed using ICP-MS. Dissolved As ranging from 4.650 to 36.894 $\mu\text{g L}^{-1}$ while dissolved Hg ranging from BDL to 0.011 $\mu\text{g L}^{-1}$. Particulate As and Hg varied from 0.650 to 86.087 mg L^{-1} and BDL to 5.873 mg L^{-1} , respectively. Higher concentration of the dissolved elements were found mainly in October 2012 and particulate elements concentration mostly higher in May 2012. The source of the studied metals in the river may be the run-off from the effluent discharges and other natural sources. The toxic elements studied in Sungai Kuantan waters were still below the Interim Marine Water Quality Standard (INWQS) permissible limits.

Keywords: Arsenic; dissolved metals; mercury; river water; suspended particulate matter

ABSTRAK

Pengawasan kepekatan logam toksik di Sungai Kuantan adalah penting kerana sungai tersebut menjadi tumpuan komuniti daripada segi domestik, perikanan dan pertanian. Oleh itu, untuk mengenal pasti logam As dan Hg dalam fasa terlarut dan pepejal terampai yang terdapat di Sungai Kuantan, sampel air di bahagian permukaan air dan di bahagian dasar telah diambil di 9 stesyen bermula dari muara sungai hingga ke hulu sungai pada Mei 2012 sehingga Oktober 2012. Logam terlarut telah dipekatkan menggunakan Chelex-100 manakala logam pepejal terampai dianalisis melalui proses penghadaman Teflon Bomb. Logam surih kemudiannya dianalisis melalui Instrumen Spektrometri Jisim Gandingan Plasma Teraruh (ICP-MS). Logam As terlarut berjangka daripada 4.650 hingga 36.894 $\mu\text{g L}^{-1}$ manakala Hg terlarut berjangka daripada BDL hingga 0.011 $\mu\text{g L}^{-1}$. Logam As dan Hg fasa pepejal terampai masing-masing berjangka daripada 0.650 hingga 86.087 mg L^{-1} dan BDL hingga 5.873 mg L^{-1} . Kepekatan unsur logam terlarut adalah tinggi pada Oktober 2012 dan logam pepejal terampai yang diperolehi adalah tinggi pada Mei 2012. Logam toksik di Sungai Kuantan berada di bawah paras yang digariskan oleh Piawai Interim Kualiti Air Marin (IMWQS).

Kata kunci: Air sungai; arsenik; logam pepejal terampai; logam terlarut; merkuri

INTRODUCTION

Toxic elements such as cadmium (Cd), arsenic (As) and mercury (Hg) cannot be decomposed or destroyed in environment that indicates they are one of conservative pollutants. These have received considerable attention because of the inherent toxicity to the living aquatic forms. The toxicity of heavy metals in water is affected by pH, hardness, alkalinity and organic material (Govindasamy et al. 1999). The presence of toxic metals in the aquatic ecosystem has far-reaching effects directly to the biological organisms and indirectly to human (Awofolu et al. 2004).

In recent years, the contamination of aquatic systems has become a problem of great concern throughout the world. Reclamation of river water for industrial, urban and other forms of development has been increasing

and it causes irreversible damage to the river and their surroundings throughout the tropics. Information on the level of toxic elements in the river of Malaysia is rare and inadequate to a few studies. Moreover, research on dissolved and particulate metals in river water is scarce due to their trace-level existence (sub-ppb level) and special precautions from sample collection to the analysis process of river water are necessary (Hunter et al. 1997). Environmental pollution monitoring is essential at strategic point in natural water bodies, as well as in commercial and industrial locations. Sungai Kuantan serves many communities in terms of domestic and agricultural purposes and it is important to carry out such an investigation. Therefore, the aims of this study were to determine the distributions of dissolved and particulate

As and Hg and evaluate its changes in Sungai Kuantan spatially and temporally.

MATERIALS AND METHODS

SAMPLING SITES

Sungai Kuantan basin covers an area of 1586 km² and located adjacent to the Tanjung Lumpur mangrove in Kuantan (DOE 2010). The sampling was carried out approximately 20 km long from the estuary to the upstream of the river. Bottles sample were rinsed thoroughly with river water from the sampling station before it was filled with the river water samples. Surface (0.3 m from the surface) and bottom (0.3 m from the bottom) water samples were taken once a month from 9 sampling points (Figure 1) from May to October 2012 using the Van-Dorn Water Sampler. The samples were stored in acid-washed polyethylene bottles (1L) and were kept in ice during the transportation to the laboratory (APHA 1998). Concomitantly, physico-chemical parameters of the water were measured using Hydrolab.

ANALYTICAL METHODS FOR TOXIC METALS

All apparatus such as plastic bags and polyethylene bottles that were used in the laboratory and during the sampling were immersed in nitric acid (HNO₃) to prevent any impurities. Water samples were filtered through dry pre-weighed Nylon millipore filters (0.45 μm pore size) with an aid of low-pressure vacuum pump (Beltrame et al. 2009). The phase that retained on the membranes was classified as particulate phase, whereas the water samples that passed through the membranes were labeled as dissolved phase.

Then, with peristaltic pump at the most optimum flow rate of 2 mL/min (Pai 1988), HNO₃, distilled water, ammonia and followed by distilled water again were passed through the column to remove all the excess that might left. The column was buffered by passing through ammonium acetate to pH5.4 (Pai 1988) and the water samples was loaded onto the Chelex-100 column and then were eluted with 5 mL 2M HNO₃. All procedures were done in a class-100 flow hood. The recovery test was also conducted through the spiking method analysis (Gebremedhin & Berhanu 2015) and the percentage of recovery ranged from 95% to 105%.

Filter papers that have dried up to constant weight were re-weighed. The filters digestion methods were followed the procedure used by Kamaruzzaman (1999). In the Teflon bomb, filters were added together with 1.5 mL of mixed acid (ratio of 3.5 HNO₃: 3.5 Hydrochloric acid (HCl): 3 Hydrofluoric acid (HF)) in a Teflon vessel and heated for 5 to 7 h at 150°C in the microwave oven. After cooling session, digested sample were added with mixed of boric acid (H₃BO₃) and EDTA and were heated again. Samples digestion was carried out in duplicate to ensure the reproducibility of the method. Percentage recoveries for metals analyses were between 95 and 110%. The toxic elements in dissolved and particulate samples were detected using Inductively Coupled Plasma Mass Spectrometer (ICP-MS).

RESULTS AND DISCUSSION

PHYSICAL PARAMETERS

The range of river water temperature in these sampling sites seemed normal for the equatorial climate (McGinley

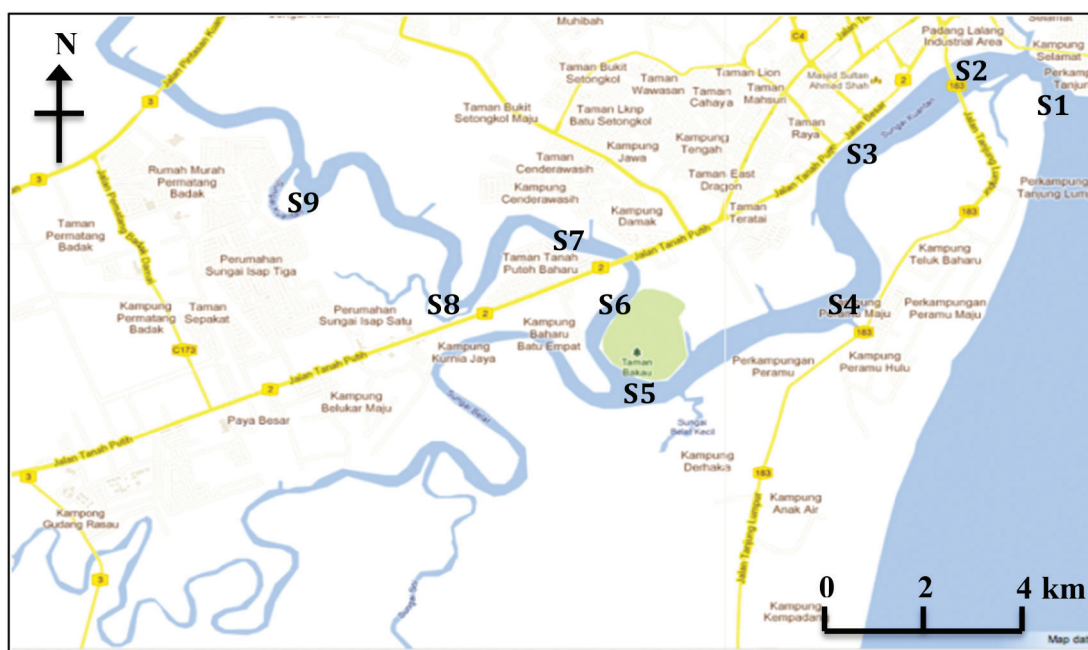


FIGURE 1. Location of sampling sites for the Sungai Kuantan

2011), ranging from 27.242 to 31.731°C. The average of salinity, pH, dissolved oxygen and precipitation were recorded in the Table 1.

DISSOLVED TOXIC METALS

Summary statistic of concentrations of dissolved As and Hg from May 2012 till October 2012 at Sungai Kuantan basin are listed in Table 1. Throughout the sampling periods, relatively higher mean concentrations of dissolved As with value of $24.101 \pm 7.792 \mu\text{g L}^{-1}$ (range: 10.400 - 38.257 $\mu\text{g L}^{-1}$) and $23.704 \pm 7.117 \mu\text{g L}^{-1}$ (range: 11.450 - 39.864 $\mu\text{g L}^{-1}$) were found in October and September 2012, respectively. The lowest mean concentration with value of $12.838 \pm 5.164 \mu\text{g L}^{-1}$ (range: 4.650 - 25.613 $\mu\text{g L}^{-1}$) was obtained in June 2012. Temporal and spatial variations of dissolved As concentration were depicted in the Figure 2. Concentrations of dissolved Hg presented were lower than

As with values below $0.015 \mu\text{g L}^{-1}$ at all sampling sites in the Sungai Kuantan basin. Mean concentration of dissolved Hg was higher during the pre-monsoon season by which the highest concentration with mean value of $0.006 \pm 0.002 \mu\text{g L}^{-1}$, ranged from below the detection limit (BDL) to $0.0111 \mu\text{g L}^{-1}$ was found in October 2012. Meanwhile, the lowest concentration of Hg that has varied between BDL and $0.004 \mu\text{g L}^{-1}$ (range: $0.001 \pm 0.000 \mu\text{g L}^{-1}$) was found in July 2012. Temporal and spatial variations of dissolved Hg concentration were depicted in the Figure 3.

Generally, Pearson correlation analysis in dissolved As and Hg showed positive and significant correlation with DO ($p < 0.01$). Temperature and pH showed negative and significant correlation with dissolved As and Hg. Negative and significant correlation was also found between salinity and As and no correlation with Hg (Table 2).

The temporal and spatial patterns of dissolved As and Hg exhibited similarity, in which higher in September and

TABLE 1. Summary statistic of dissolved and particulate As and Hg concentration and parameters of Sungai Kuantan basin

Element		May 2012	June 2012	July 2012	August 2012	September 2012	October 2012
Dissolved As ($\mu\text{g L}^{-1}$)	Average	13.882	12.837	15.568	20.217	23.703	24.100
	SD	4.149	5.164	5.585	6.220	7.116	7.791
	Min	6.398	4.650	5.250	8.111	11.449	10.400
	Max	23.016	25.613	28.094	30.997	39.864	38.257
Particulate As (mg L^{-1})	Average	36.310	34.522	27.602	20.236	26.189	19.304
	SD	22.841	18.073	16.509	15.532	12.528	12.747
	Min	3.236	1.463	5.212	0.650	6.403	1.494
	Max	86.087	84.180	68.629	66.507	55.915	49.369
Dissolved Hg ($\mu\text{g L}^{-1}$)	Average	0.482	0.342	0.273	1.121	1.884	2.036
	SD	0.127	0.152	0.157	1.071	1.193	1.897
	Min	BDL	BDL	BDL	BDL	BDL	BDL
	Max	2.392	1.727	1.278	4.098	5.873	5.609
Particulate Hg (mg L^{-1})	Average	0.482	0.342	0.273	1.121	1.884	2.036
	SD	0.127	0.152	0.157	1.701	1.193	1.897
	Min	BDL	BDL	BDL	BDL	BDL	BDL
	Max	2.392	1.727	1.278	4.098	5.873	5.609
Salinity (ppt)	Average	21.450	27.430	26.630	22.140	22.360	20.080
	SD	9.900	5.960	6.000	10.870	9.910	10.600
	Min	0.330	190	11.820	1.290	3.510	0.090
	Max	34.300	34.770	34.380	34.410	33.960	33.570
pH	Average	7.110	7.960	7.910	7.150	6.890	6.550
	SD	0.580	0.800	0.840	0.450	0.460	0.500
	Min	6.660	7.060	6.820	6.280	6.120	6.020
	Max	7.820	8.970	8.620	8.010	7.520	7.430
DO (mg/L)	Average	5.400	5.070	4.910	5.140	5.660	6.250
	SD	0.640	0.500	0.460	0.670	0.840	0.800
	Min	4.460	3.540	3.730	4.020	4.120	4.710
	Max	6.610	5.950	5.790	6.220	6.980	7.670
Temperature ($^{\circ}\text{C}$)	Average	29.790	30.220	30.670	29.850	29.290	28.920
	SD	0.630	0.280	0.660	0.400	0.900	1.010
	Min	28.150	29.800	29.850	29.020	27.360	27.242
	Max	30.780	31.150	31.731	30.240	30.300	30.050
Precipitation (mm) (DOE, 2013)	Average	146.6	103.2	166.8	181.6	218.2	288.6

BDL = Below Detection Limit

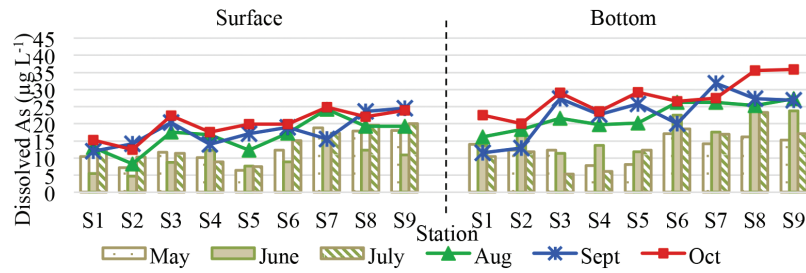


FIGURE 2. Concentrations of dissolved As ($\mu\text{g L}^{-1}$) at different sampling periods

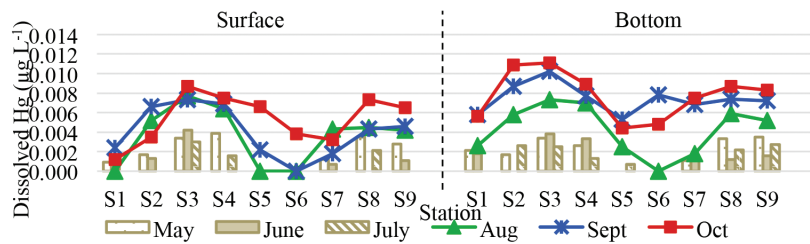


FIGURE 3. Concentrations of dissolved Hg ($\mu\text{g L}^{-1}$) at different sampling periods during high tide

October 2012, during the onset of the monsoon season which high precipitation recorded. Dissolved As and Hg levels increased when the precipitation increased, probably related to the higher tidal currents, which induce re-suspension of particles and mobilization of pore water in the shallow and generally, more contaminated areas (Ramalhosa et al. 2006a). In addition, the high concentration of dissolved As may direct to the possibility of some As associated with iron as FeAsS, leaching into river and of the dissolution of river-borne sediments (Nair et al. 2003).

Lower concentration of dissolved As and Hg in May 2012 during the lower discharge possibly caused by slow water flow conditions that bring to minimal influx of freshwater thus enhanced sedimentation. Sedimentation would likely become the more important mechanism for removing heavy metals and other pollutants from the water column especially during the low tide (Ezeronye & Ubalua 2004). Low levels of Hg at the estuary possibly caused by the non-conservative removal exhibited by dissolved Hg along the salinity gradient, which can be associated to flocculation and particle scavenging through sorption and co-precipitation processes (Han et al. 2006). Dissolved Hg elevated at the S3 and S4 in accordance with its location at the city centers which in their massive development. Rainfall acts as an external source by increased land runoff and washed down the particulate matter to the water column. Positive and significant correlations between As and Hg shown in Table 3 indicates both elements might possess high similarities in the sources, environmental distribution and chemical behavior.

Moreover, dissolved As average concentrations was measured to be lower than the other river waters in Malaysia

such as Sungai Semenyih ($< 1000.000 \mu\text{g L}^{-1}$) and Sungai Lembing ($40.000 \mu\text{g L}^{-1}$). No water sample registered As and Hg concentrations exceeding the INWQS (DOE 2013) Class II of $50 \mu\text{g L}^{-1}$ and $10 \mu\text{g L}^{-1}$, respectively and no sampling location registered dissolved Hg concentrations above the WHO (2006) drinking water guideline value of $6.000 \mu\text{g L}^{-1}$ and Criteria Max Concentration (EPA 2004) guideline value of $1.400 \mu\text{g L}^{-1}$.

PARTICULATE TOXIC METALS

Particulate As showed concentrations ranging from 0.650 to 86.087 mg L^{-1} in Sungai Kuantan. Throughout the sampling periods, a relatively higher mean concentrations of particulate As with mean value of $36.310 \pm 22.841 \text{ mg L}^{-1}$ (range: $3.236 - 86.087 \text{ mg L}^{-1}$) was found in May 2012 while October 2012 showed the lowest mean concentration with value of $19.304 \pm 12.747 \text{ mg L}^{-1}$ (range: $1.494 - 49.369 \text{ mg L}^{-1}$). Temporal and spatial variations of particulate As are depicted in Figure 4. The highest concentration with mean value of particulate Hg was $2.036 \pm 1.897 \text{ mg L}^{-1}$, ranged from BDL to 5.609 mg L^{-1} found in October 2012 while the lowest particulate Hg concentration that varied between nd and 1.278 mg L^{-1} with the corresponding mean value of $0.273 \pm 0.157 \text{ mg L}^{-1}$ was obtained in July 2012. Temporal and spatial variations of particulate Hg are depicted in Figure 5. Pearson correlation in particulate fractions showed that salinity, temperature and pH were negative and significantly correlated with particulate As and Hg (Table 2).

High flux values due to higher rainfall corresponded to lower concentration of particulate As, whereas low fluxes with small volume corresponded to higher concentration. Plamateer et al. (1993) suggested that, generally, SPM size

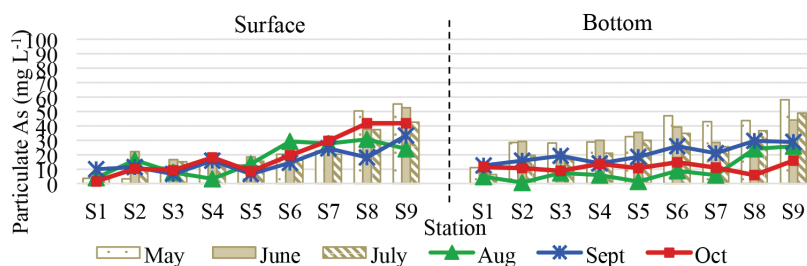


FIGURE 4. Concentrations of particulate As (mg L^{-1}) at different sampling periods

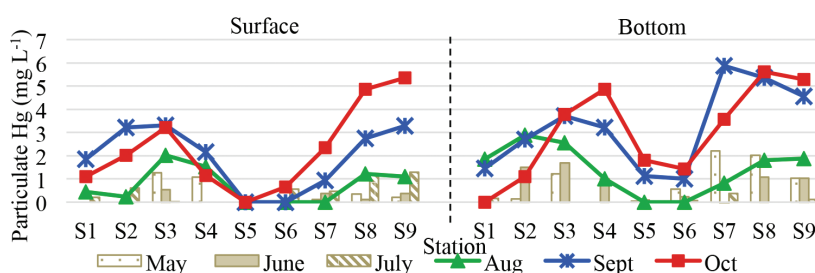


FIGURE 5. Concentrations of particulate Hg (mg L^{-1}) at different sampling periods

TABLE 2. Correlation analysis of physico-chemical parameters with studied elements

	Salinity	Temperature	pH	DO
Dissolved As	-.554**	-.640**	-.600**	.462**
Dissolved Hg	-.119	-.346**	-.442**	.306**
Particulate As	-.475**	-.266**	-.203**	-.051
Particulate Hg	-.299**	-.637**	-.408**	.568**

**Correlation is significant at the 0.01 level (2-tailed)

*Correlation is significant at the 0.05 level (2-tailed)

ranges from 0.1 to 200 μm . Particles above this size could be introduced and transported during high flows, but during lower flows, the rivers would not have enough energy to transport this particle size (Dickinson 1987). This statement could explain an observed decrease in metal concentration during some of the highest flow peaks that mostly occurred during the rainy season. Cardoso Fonseca and Ferreira da Silva (2000) claimed that the amorphous and crystalline Fe oxides and also the sulphides were the main bearing-phases of As. Thus the present of those metals might elevate the As levels in Sungai Kuantan. Particulate As also may release during steel production at the industrial sites located at the upper stream station (S8 and S9) (Ondov et al. 1996).

Concentrations of particulate Hg were fluctuated along the flow path towards the upstream stations. Variations in particulate load in the water column were a result of higher fresh water input (rivers and land drainage), in addition to wave action and wind. Fluctuations of Hg in SPM concentrations are essentially a function of current velocity and a result of erosion/deposition processes, which may also be induced by the wind. In addition, the occurrence of re-suspension related to wind-induced turbulence can also

contribute to the increase of bed-shear stress mediated by wave-induced sediment stirring (Paphitis & Collins 2005) and change the distribution of suspended particles during the tidal cycles as well as higher particulate Hg levels at the bottom layer of the water column. Other studies by Choe et al. (2003), Kim et al. (2004) and Schäfer et al. (2006) also observed the importance of re-suspension as a source of particulate mercury in the water column. Sungai Kuantan also registered Hg concentration above the International Commission for the Protection of the Rhine against Pollution (ICPR) Hg limit of 0.500 mg L^{-1} (ICPR 1991).

A good positive correlation that was found between particulate As and Hg shows that there were common source or at least one major source for all of them and may reflects similar behavior (Table 3). Zhou et al. (2003) whom also found significant inter-element relationships in SPM claimed that some trace metals have similar reactivity towards biological and non-biological particle and therefore possible for some of them to have their particulate-bound concentrations closely related to each other.

TABLE 3. Pearson's correlation matrix indicating the overall metals association within dissolved elements

	Dissolved As	Dissolved Hg	Particulate As	Particulate Hg
Dissolved As	-	.465**	-.164*	.428**
Dissolved Hg	-	-	.368**	.368**
Particulate As	-	-	-	0.39*

**Correlation is significant at the 0.01 level (2-tailed)

*Correlation is significant at the 0.05 level (2-tailed)

CONCLUSION

Concentrations of dissolved As, particulate As and dissolved Hg were found higher during the rainy season with mean value of $24.101 \pm 7.792 \mu\text{g L}^{-1}$, $0.006 \pm 0.002 \mu\text{g L}^{-1}$ and $36.310 \pm 22.841 \text{mg L}^{-1}$, respectively, while particulate Hg was higher during the dry season with mean value of $2.036 \pm 1.897 \text{mg L}^{-1}$. Both dissolved As and Hg concentrations were below the INWQS permissible limits. Heavy rainfall and higher river flow may lead to re-suspension of bed sediments enriched with trace metals. Concentrations in dissolved and particulate metals increasing from downstream to the upstream station shows that metal concentration inverse with the salinity. Heavily urbanized city centers nearby the river may contribute to the significant amounts of metals through land run-off, effluents and waste from the drains.

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