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Source profiling of arsenic and heavy metals in the Selangor River basin and their maternal and cord blood levels in Selangor State, Malaysia

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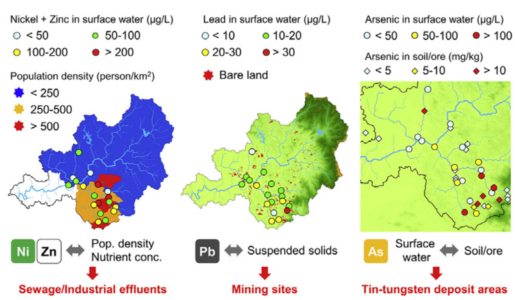
HIGHLIGHTS

- Source profiling of As and 5 heavy metals was performed in the Selangor River basin.
- Maternal and cord blood levels of As, Cd and Pb were analyzed in 136 pregnant women.
- Ni and Zn derived from sewage/industrial effluents, while Pb derived from mining.
- As showed a site-specific pollution from soil/ore in tin-tungsten deposit areas.
- The maternal and cord blood levels of As, Cd and Pb were not acute toxicity levels.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic and 5 heavy metals (nickel, copper, zinc, cadmium and lead) were quantitated in surface water ($n = 18$) and soil/ore samples ($n = 45$) collected from 5 land uses (oil palm converted from forest, oil palm in peat swamp, bare land, quarry and forest) in the Selangor River basin by inductively coupled plasma mass spectrometry (ICP-MS). Geographic information system (GIS) was used as a spatial analytical tool to classify 4 land uses (forest, agriculture/peat, urban and bare land) from a satellite image taken by Landsat 8. Source profiling of the 6 elements was conducted to identify their occurrence, their distribution and the pollution source associated with the land use. The concentrations of arsenic, cadmium and lead were also analyzed in maternal blood ($n = 99$) and cord blood ($n = 87$) specimens from 136 pregnant women collected at the University of Malaya Medical Center for elucidating maternal exposure as well as maternal-to-fetal transfer. The source profiling identified that nickel and zinc were discharged from sewage and/or industrial effluents, and that lead was discharged from mining sites. Arsenic showed a site-specific pollution in tin-tungsten deposit areas, and the pollution source could be associated with arsenopyrite. The maternal blood levels of arsenic (0.82 ± 0.61 μg/dL), cadmium (0.15 ± 0.2 μg/dL) and lead (2.6 ± 2.1 μg/dL) were not significantly high compared to their acute toxicity

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levels, but could have attributable risks of chronic toxicity. Those in cord blood were significantly decreased in cadmium (0.06 ± 0.07 μg/dL) and lead (0.99 ± 1.2 μg/dL) but were equivalent in arsenic (0.82 ± 1.1 μg/dL) because of the different kinetics of maternal-to-fetal transfer.

1. Introduction

The Selangor River basin is located in the northern part of the capital region in Malaysia. Approximately 60% of water supply in the capital region is derived from the Selangor River basin (Manap et al., 2013), and the rest is derived from the Klang and the Langat River basins, which are located in the center and southern parts of the capital region, respectively. Deterioration in water quality has been a concern for these river basins because drinking water treatment facilities have been shut down frequently due to a high concentration of ammonia in intake water. The concentration is associated with the acceleration of economic growth (Hasan et al., 2010; Juahir et al., 2011). Serious pollution of arsenic and heavy metals in the capital region has also been reported in recent years. For instance, high concentrations of zinc (52.9 μg/L), arsenic (24.7 μg/L) and lead (8.0 μg/L) have been detected in surface water collected at the estuary of the Klang River basin (Sany et al., 2013). The arsenic concentration exceeded a recommended raw water quality standard (10 μg/L) set by the Ministry of Health Malaysia (2009). However, few reports have analyzed arsenic and heavy metals in the Selangor River basin compared to the Klang and Langat River basins, although the former is the largest water resource in the capital region.

The Selangor River basin used to be one of the most active tin-mining areas in Peninsular Malaysia (Schwartz et al., 1995). Tin-mining sites are usually enriched by not only tin but also arsenic and other heavy metals. Soils near former tin-mining ponds have been found to contain tin (404 mg/kg), copper (125 mg/kg), zinc (115 mg/kg), lead (105 mg/kg) and arsenic (79 mg/kg) (Ashraf et al., 2011). Sediments at former tin-mining ponds have also been found to contain tin (100–112 mg/kg), zinc (62.4–72.0 mg/kg), lead (26.1–32.4 mg/kg), copper (7.7–22 mg/kg) and arsenic (0.11–0.23 mg/kg) (Ashraf et al., 2012). The local authority has proposed to supply alternative raw water from abandoned mining ponds combined with groundwater abstraction, a proposal that is called a hybrid off-river augmentation system (HORAS) and is intended to mitigate the severe water shortage experienced in recent years. The project started with the implementation of water pumping from abandoned mining ponds into the Selangor River in 2014 (Kusin et al., 2016). Extensive natural forests in the Selangor River basin were converted into agricultural land, mainly for rubber and oil palm plantations from the 1950s to the 1970s, and the forest fragmentation has been accelerated by land development for housing, new urban areas and industrial estates since the 1980s (Abdullah and Nakagoshi, 2007). Consequently, the surface water has been heavily contaminated by *Escherichia coli* (*E. coli*), nutrients and suspended solids, particularly in populated areas (Othman et al., 2014). Therefore, the contamination of arsenic and heavy metals as well as their pollution sources in the Selangor River basin need to be elucidated.

The concentrations of arsenic and heavy metals such as cadmium and lead in freshwater and marine species on the west coast of Peninsular Malaysia exceed permissible levels, as established by the Malaysian Food Regulations 1985 (Ministry of Health Malaysia, 1985; Yap et al., 2002; Cheng and Yap, 2015). Arsenic concentrations in groundwater collected nationwide frequently exceed national drinking water quality standards (10 μg/L) (Department of Environment Malaysia, 2015). Therefore, local people could be significantly exposed to arsenic and heavy metals via food and water consumption. However, few reports have analyzed them in the Malaysian population, and their blood levels in pregnant women have not been elucidated. Maternal exposure to arsenic and heavy metals is crucial to fetuses because placentas cannot completely prevent maternal-to-fetal transfer (Korpela et al., 1986).

This study focused on arsenic and 5 heavy metals (nickel, copper, zinc, cadmium and lead) and quantitated them in surface water and soil/ore samples collected in the Selangor River basin by inductively coupled plasma mass spectrometry (ICP-MS). Hydrochloric acid was used for extracting labile forms of metals in soil/ore samples, such as metals from iron and manganese oxides or organic phases (Snape et al., 2004), that could be associated with those detected in the surface water. Source profiling of the 6 elements was conducted to elucidate their occurrence, their distribution and the pollution source using geographic information system (GIS). Three elements (arsenic, cadmium and lead) in maternal and cord blood specimens of local pregnant women, collected at the University of Malaya Medical Center, were also analyzed to elucidate maternal exposure and the maternal-to-fetal transfer. Maternal blood levels were compared with toxicological studies to assess the potential health risks. Additionally, the exposure pathways via food and water consumption were investigated based on a literature review. Thus, this study aimed to elucidate the occurrence and distribution of arsenic and heavy metals in the Selangor River basin and to assess their health risks to pregnant women in Selangor State, Malaysia.

2. Materials and methods

2.1. Sample collection and water quality monitoring

The sampling sites for surface water and soil/ore in the Selangor River basin are described in Fig. 1. The sampling sites for surface water (n = 18) were selected mainly from the Selangor River (S13) and its tributaries (S1-S12), where the surface water quality is deteriorated by anthropogenic influence (Fulazzaky et al., 2010). A small tributary along a peat swamp (S16 and S17) was selected because of the unique characteristics of its dark brown color and low pH (Sakai et al., 2016). Three points in the main stream (S14, S15 and S18) were also selected to monitor the transition of water quality and heavy metal contamination. Soil and ore samples were collected at oil palm converted from forest, oil palm in peat swamp, bare land, quarry (*e.g.,* limestone, bauxite, sand and granite) and forest near the sampling sites of surface water. Surface soils (0–5 cm) were collected at oil palm converted from forest, bare land and forest, and core samples (0–110 cm in depth) were collected at oil palm in peat swamp (Table S1). All the samples (n = 45) were collected during 1 week from July 19th to 26th, 2015.

The surface water was collected using a stainless steel container, from which one liter was poured into a polypropylene bottle. All the samples (n = 18) were collected on July 23rd, 2015 when there was no precipitation. During the sampling, the pH and electrical conductivity (EC) were measured by LAQUAtwin (Horiba, Japan); the