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

Nobumitsu Sakai¹,², *, Zohour Alsaad¹, Nguyen Thi Thuong³, Kenji Shiota⁴, Minoru Yoneda⁴, Mustafa Ali Mohd⁵

¹ Division of Environmental Engineering, Graduate School of Engineering, Kyoto University, Kyoto, 6158540, Japan
² Shimadzu-UMMC Centre of Xenobiotic Studies, Department of Pharmacology, Faculty of Medicine, University of Malaya, Kuala Lumpur, 50603, Malaysia

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 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.

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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 (Fulazakzky 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 LAQUAwin (Horiba, Japan); the dissolved oxygen was measured using an Accumet AP84 (Fisher Scientific, Malaysia); and the river width, water level and velocity were measured to estimate a flow rate (Table S2). The collected samples were placed in a cooler, transferred to the laboratory and stored at 4 °C until further analysis.

Ammonia nitrogen (NH₄-N) and Escherichia coli (E. coli) in the surface water samples were analyzed by a digital pack test apparatus (Digital Pack Test Multi, Kyoritsu Chemical-Check Lab. Corp., Japan) and Colilert method (IDEXX Laboratories Inc., USA), respectively. Suspended solids (SS) were analyzed by filtering the surface water sample through a GF/B glass microfiber filter (GE Healthcare UK Ltd., UK) and drying the filter at 105 °C for 2 h. The pH and EC

![Fig. 1. Location of surface water (S1-S18; n = 18) and soil/ore (n = 45) samples in the Selangor River basin. The soil/ore samples were collected at 5 land uses (oil palm converted from forest, oil palm in peat swamp, bare land, quarry and forest). Geographic information for sampling sites is presented in Table S1.](image-url)
in the soil/ore samples mixed with pure water were analyzed by LAQUAtwin (Horiba, Japan), and the moisture content was measured by comparing a weight before and after completely drying at 105 °C overnight (Table S3).

2.2. Sample pretreatment and ICP-MS analysis

Twenty milliliters of surface water samples were mixed with 0.4 mL of nitric acid (65%) diluted by pure water (1:1 v/v) and were digested at 80 °C for 2 h. No filtration before the acid digestion was made to analyze the concentrations of arsenic and heavy metals in the surface water including suspended solids to elucidate the pollution source associated with the soil/ore. The aliquot was filtered through a 0.2 μm regenerated cellulose syringe filter (Phenex, Phenomenex Inc., USA) and diluted 20 times by pure water for adjusting to the detection range of ICP-MS. One gram of soil/ore samples was mixed with 35 mL of hydrochloric acid (1 mol/L) and shaken at 200 rpm for 2 h to extract labile and weakly sorbed metals. After stabilizing for 30 min, the aliquot was filtered through a 0.2 μm regenerated cellulose syringe filter and diluted 50 times by pure water for adjusting to the detection range of ICP-MS. Six elements (nickel, copper, zinc, arsenic, cadmium and lead) in the final solution were analyzed by ICP-MS (X Series 2, Thermo Scientific, USA). No internal standard was used and the 6 elements were quantified by an external standard method using multielement standard solutions (W-V and W-X, Wako, Japan). Those which were quantified less than a limit of detection (nickel: 0.003 μg/L, copper: 0.044 μg/L, zinc: 0.141 μg/L, arsenic: 0.013 μg/L, cadmium: 0.013 μg/L and lead: 0.004 μg/L) in the final solutions were shown as not detected (ND).

2.3. Spatial analysis by GIS

Geographic information system (ArcGIS version 10.0, ESRI Inc., USA) was used for spatial analysis and source profiling. Digital elevation data (3sec GRID; Conditioned DEM in HydroSHEDS) were downloaded from the United States Digital Service (http://hydrosheds.cr.usgs.gov/dataavail.php), and a watershed boundary was visualized with ArcGIS Spatial Analyst for the Selangor River basin and 18 catchment areas corresponding to the sampling sites. The population distribution dataset for Malaysia at a resolution of approximately 1 km (LandScan 2012) was purchased from the Oak Ridge National Laboratory. The population density in the 18 catchment areas was calculated based on the population within the boundaries (Fig. S1). Landsat 8 OLI images at the study area captured on May 30th, 2015 were downloaded from the United States Geological Survey (http://earthexplorer.usgs.gov/). A true color image created by its bands 2, 3 and 4 was categorized by 3 land uses (forest, agriculture/peat and urban) with Iso Cluster unsupervised classification. The unsupervised classification was generalized using ArcGIS Spatial Analyst tools, such as the majority filter, boundary clean and nibble functions (Langford, 2007). Furthermore, bare land areas in the whole river basin were manually drawn from the true color image, and its layer was created by GIS for the precise extraction of bare land areas. This procedure was used because urban and bare land areas could not be differentiated sufficiently by the unsupervised classification due to a number of buildings that had a brightly colored roof similar to the color of bare land (Fig. S2). The manual extraction was validated both with high-resolution satellite images in Google Earth Pro and with our field survey data. The unsupervised classification layer was overlaid with the bare land layer to create a land-use map with the 4 categories. The overlaid ratio between the urban area in the unsupervised classification and the bare land layer was calculated at 92.2%, which means that most of the bare land areas could be differentiated from the urban area.

2.4. Statistical and spatial analysis for source profiling of arsenic and heavy metals

A correlation coefficient between NH_4-N concentration and population density among 17 sites (all except S18) were calculated to elucidate the nutrient levels associated with population. A correlation coefficient between daily load of SS and bare land areas was also calculated among the 17 sites. S18 was excluded because it was located downstream from 3 water intake points (Sungai Selangor Phase 1, 2 and 3), where a substantial amount of surface water was abstracted (Kusin et al., 2016; Fulazzaky, 2013). Bivariate and factor analyses among the 6 elements, NH_4-N and SS concentrations and the population density were analyzed by IBM Statistical Package for the Social Sciences (SPSS Version 21, IBM, USA) to identify their pollution sources based on the relationships. Furthermore, the concentrations of the 6 elements in soil/ore samples (n = 45) among the 5 land uses (oil palm converted from forest, oil palm in peat, bare land, quarry and forest) and those in surface soils (0–50 cm, n = 21) and deep soils (>50 cm, n = 8) collected at oil palm converted from forest and oil palm in peat were compared to identify the potential for arsenic and heavy metal contamination from these land uses. The occurrence and distribution of arsenic were spatially assessed by plotting its concentrations in both surface water and soil/ore samples on the watershed map to identify a site-specific contamination.

2.5. Analysis of arsenic, cadmium and lead in maternal and cord blood

Maternal blood (n = 99) and cord blood (n = 87) specimens of 136 pregnant women living in Selangor State (paired specimens: 50 subjects; unpaired specimens: 86 subjects) were collected at the University of Malaya Medical Center from January to September 2014. Of the 136 newborns, 109 newborns were born by normal delivery and 27 newborns were delivered by lower segment cesarean section or vacuum extraction. Most newborns were in normal condition, although 10 newborns were delivered before 37 weeks and 4 newborns had a low Apgar score (<7). They were immediately put into EDTA tubes (BD Vacutainers, Becton Dickinson, USA) and stored at −80 °C until further analysis. Each specimen (0.2 mL) was diluted by 8.8 mL of pure water, and 1 mL of nitric acid and 0.1 mL of indium solution (100 μg/L) for an internal standard were added. The aliquot was filtered through a 0.45 μm cellulose acetate membrane filter (Advantage, Japan), and 3 elements (arsenic, cadmium and lead) in the final solution were analyzed by ICP-MS with an internal standard method. The others (nickel, copper and zinc) were not analyzed because of their relatively less toxicity levels compared to arsenic, cadmium and lead.

3. Results and discussion

3.1. Water quality and anthropogenic impacts in the Selangor River basin

There was a moderate correlation (r = 0.53) between the NH_4-N concentration and the population density (Fig. 2). The NH_4-N concentrations in most of populated areas (more than 250 person/km²) exceeded 1 mg/L, whereas the NH_4-N concentrations in less populated areas such as S14-S18 (less than 250 person/km²) were below 1 mg/L (Fig. 2 and Table S2). Additionally, the EC and E. coli
showed strong positive correlations with NH$_4$-N ($r = 0.89$, and 0.86, respectively), while the DO showed a strong negative correlation ($r = -0.83$) with NH$_4$-N (Table S2). There was a strong correlation ($r = 0.95$) between the daily load of SS and bare land area (Fig. 3). The SS concentration at S10 was extremely high (2450 mg/L) because of mining activities nearby, and the daily load of SS was mainly produced from S4, S9 and S10 and transported toward S11 and S13 (Table S2). S15 received 252 ton/day of SS from upstream; however, the daily load decreased to 9.6 ton/day at S18 because a substantial amount of surface water was abstracted at 3 water intake points (Kusin et al., 2016). These correlations clearly suggest that the high NH$_4$-N, EC and $E_{coli}$ concentrations as well as the low DO concentration were due to untreated municipal wastewater or intermittent unsatisfactory discharges from combined sewer overflows (Rathnayake and Tanyimboh, 2015) and that the high SS loads were due to mining operations, housing and road development, logging and forest clearing (Mohamed et al., 2015).

3.2. Source profiling of arsenic and heavy metals

Four elements (nickel, zinc, arsenic and lead) were detected at all sampling points of surface water, whereas copper was detected at 3 sites and cadmium was not detected at all (Table S4). Some significant correlations were found between the nickel and zinc concentrations ($P < 0.1$), nickel and NH$_4$-N concentrations ($P < 0.1$), zinc and NH$_4$-N concentrations ($P < 0.01$), zinc concentration and population density ($P < 0.01$) and lead and SS concentrations ($P < 0.01$) (Table 1). These correlations suggest that nickel and zinc were mainly dis-

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**Fig. 2.** Spatial distribution of population densities analyzed in 18 catchment areas (left) and a correlation coefficient between population density and NH$_4$-N concentration in surface water in 17 sites except S18 (right). The spatial distribution of population in the Selangor River basin, which was used for calculating the density of population as well as catchment areas of the 18 sampling sites, are presented in Fig. S1.

**Fig. 3.** Land use in the Selangor River basin analyzed by unsupervised classification with GIS (left) and a correlation coefficient between total bare land area and the daily load of SS in 17 catchment areas except S18 (right). A true color image of Landsat 8 (bands 2, 3 and 4) captured on May 30th, 2015 was used for the unsupervised classification. The classified image was categorized by 3 land uses (forest, agriculture/peat and urban) and overlaid with a bare land layer, which was manually extracted from the true color image to create a land use map with the 4 categories. The true color image and the bare land layer are presented in Fig. S2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
charged from sewage or industrial effluents because the high contamination sites such as S5 and S12 were located at populated areas (Fig. 2). Lead was predominantly produced from mining activities because S7 was located near a mining site and showed a significantly high level of lead (329 μg/L) and SS (2450 mg/L) compared to other sites (Table S4). The factor analysis also supported these correlations from the bivariate analysis (Table 1). Principal component 1 showed a positive correlation among the nickel, zinc and NH₄-N concentrations and population density, and principal component 2 showed a positive correlation between the lead and SS concentrations. On the other hand, copper was detected only at S4, S7 and S9. The pollution source cannot be explained by population and land use because the water quality at the highest detection (S9: 74.7 μg/L) was rather clean compared to other populated areas (Table S2), and because the land use mainly consists of urban areas rather than oil palm plantation where a higher amount of copper was detected (Fig. 4). Therefore, the inconsistent detection of copper seemed to be associated with site-specific point sources that could not be identified in this study. These heavy metals are transported to the estuary, where they accumulate in sediment and marine species. Significant correlations have been found among the cadmium, copper and lead concentrations in soft tissues of green-lipped mussels and sediments in the west coast of Peninsular Malaysia (Yap et al., 2002). The nickel, arsenic, cadmium and lead concentrations at coastal areas near the capital region were far higher than their background concentrations in the west coast of Peninsular Malaysia (Sany et al., 2013; Lim et al., 2012). The cadmium and lead concentrations in green-lipped mussels (Yap et al., 2002) and arsenic concentration in mangrove snails (Cheng and Yap, 2015) in the west coast of Peninsular Malaysia exceeded the permissible levels established by the Malaysian Food Regulations 1985 (Ministry of Health Malaysia, 1985).

The concentrations of the 6 elements in soil/ore samples collected at oil palm converted from forest and oil palm in peat swamp were higher than those found at bare land, quarry and forest (Fig. 4a). The concentrations of nickel, zinc, arsenic, cadmium and lead in surface soils (0–50 cm) at oil palm converted from forest and oil palm in peat swamp were significantly higher than those found in deep soils (>50 cm) (Fig. 4b). Peat soils are generally deficient in micronutri-

![Fig. 4](image)

**Table 1**

Bivariate correlation and factor analysis among arsenic and 5 heavy metals, SS and NH₄-N concentrations in surface water and population density in catchment areas of sampling sites. Factors shown in bold in the bivariate correlation analysis are indicated as significant correlations ($P<0.01$) or trends towards significance ($P<0.1$). Factors shown in bold in the factor analysis are indicated as moderate or strong factor loadings ($>0.5$).

<table>
<thead>
<tr>
<th>Factor</th>
<th>Bivariate correlation</th>
<th>Factor analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>0.427**</td>
<td>0.534</td>
</tr>
<tr>
<td>As</td>
<td>-0.218</td>
<td>0.875</td>
</tr>
<tr>
<td>Pb</td>
<td>0.172</td>
<td>-0.010</td>
</tr>
<tr>
<td>SS</td>
<td>0.181</td>
<td>-0.004</td>
</tr>
<tr>
<td>NH₄-N</td>
<td>0.468**</td>
<td>0.874</td>
</tr>
<tr>
<td>Pop. density</td>
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<td>0.895</td>
</tr>
</tbody>
</table>

Eigenvalue

<table>
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<th></th>
<th>PC1</th>
<th>PC2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Variance (%)</td>
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<td>32.7</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>37.7</td>
<td>70.4</td>
</tr>
</tbody>
</table>

*$P<0.01$, **$P<0.1$.

Abbreviation: PC: principal component; Pop. density: population density.
ents, particularly boron, iron, copper and zinc, and the available forms of copper and zinc in peat soils change into non-available forms due to strong binding with humic and fulvic acids, thereby forming stable metal-organic matter complexes (Ambak and Tadano, 1991; Yonebayashi et al., 1994). Therefore, the heavy metals in the surface soils could be attributed to excess applications of fertilizers. The non-point source pollution of heavy metals from the oil palm plantation is a nationwide concern because approximately 12% of all land in Malaysia is covered by oil palm plantations (Wicke et al., 2011).

Arsenic showed the unique tendency of existing in higher concentrations in surface water and soil/ore samples in particular areas (Fig. 5). The surface water in S1, S6 and S8 showed more than 100 µg/L of arsenic and distributed downstream toward S4, S11 and S13. Soil/ore samples around these areas also showed higher arsenic concentrations (>5 mg/kg) compared to other sites (Table S5). Arsenic could have been deposited originally around these areas because even the quarry and forest samples, which were not influenced by anthropogenic activities, showed high arsenic concentrations. According to Schwartz et al. (1995), the high contamination areas are located at tin-tungsten deposits areas (Schwartz et al., 1995). Soils near former tin-mining ponds contained high arsenic concentrations (Ashraf et al., 2011), and tilapia in a former tin-mining pond showed higher arsenic concentrations compared to those in concrete tanks and eel pond (Low et al., 2015). In addition, a significant enrichment of arsenic in river water around gold mines in Peninsular Malaysia was observed due to arsenopyrite (Abu Bakar et al., 2015). Therefore, a high potential exists for the contamination of arsenic from natural sources in these particular areas, and mining activities occurring near S1, S6 and S7 could aggravate the pollution. Speciation of arsenic by oxidation states, which are mostly found in trivalent arsenite or pentavalent arsenate, could be useful to identify the pollution source because concentrations and relative proportions of trivalent and pentavalent arsenic in surface water vary according to changes in input sources, redox conditions and biological activity (Smedley and Kinniburgh, 2002).

Hydrochloric acid is a strong acid that helps liberate metals from iron and manganese oxides, decomposes labile organic phases as well as amorphous sulphides that control metal bioavailability in anoxic, and partially oxidizes the soil matrix (Snape et al., 2004). Extraction efficiency of metals would be influenced by the soil/ore matrix as well as experimental parameters such as acid strength, extraction time, sample to solution ratio, temperature and pressure (Snape et al., 2004). More fractions such as mobile fraction assessed by a salt solution (e.g., sodium nitrate, calcium chloride and ammonium nitrate), mobilisable fraction assessed by a complexing agent (e.g., ammonium acetate, pentetic acid and ethylenediaminetetraacetic acid) as well as pseudo total metal concentration would be useful to further specify the pollution source (Gupta et al., 1996).

3.3. Blood levels of arsenic, cadmium and lead in pregnant women

Fig. 6 shows the concentrations of arsenic, cadmium and lead in maternal blood (n = 99) and cord blood (n = 87) including unpaired subjects. An equivalent concentration of arsenic was observed in maternal blood (0.82 ± 0.61 μg/dL) and cord blood (0.82 ± 1.1 μg/dL). In contrast, the concentrations of cadmium and lead in maternal blood (0.15 ± 0.2 and 2.6 ± 2.1 μg/dL, respectively) were significantly higher than those in cord blood (0.06 ± 0.07 and 0.99 ± 1.2 μg/dL, respectively) (P < 0.05). The concentrations of lead and cadmium in maternal blood were higher than those in cord blood worldwide (Korpela et al., 1986; Osman et al., 2000; Soong et al., 1991; Ploëckinger et al., 1992; Baranowska, 1995; Sakamoto et al., 2010). High concentrations of cadmium and lead were found in amniotic fluid and amniotic membranes (Korpela et al., 1986), and their accumulation in placentas has also been reported (Baranowska, 1995). Therefore, the amniotic membranes and placentas could work as barriers for the maternal-to-fetal transfer of cadmium and lead. Of the 50

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**Fig. 5.** Spatial distribution of arsenic detected in surface water and soil/ore samples. The arsenic concentration in each soil/ore sampling site represents a highest value observed in the respective samples. The arsenic concentrations of each surface water and soil/ore sample are presented in Tables S4 and S5.

**Fig. 6.** Blood levels of arsenic, cadmium and lead in pregnant women (maternal blood: n = 99; cord blood: n = 87, including 50 paired specimens) collected at University of Malaya Medical Center (*P < 0.05).
pair subjects, there was a weak positive correlation of lead concentrations between maternal blood and cord blood \((r = 0.33)\) but not in arsenic \((r = 0.04)\) and cadmium \((r = 0.16)\) concentrations. Significant correlations between maternal and neonatal blood were reported in lead (Korpela et al., 1986; Plööckinger et al., 1992), which indicates that fetal exposure to lead strongly reflected the maternal exposure level.

The average concentrations of arsenic, cadmium and lead in maternal blood were equivalent to or relatively lower than previous reports (Korpela et al., 1986; Soong et al., 1991; Plööckinger et al., 1992; Baranowska, 1995; Sakamoto et al., 2010). The average lead concentration \((2.6 \mu g/dL)\) was over \(2 \mu g/dL\), and this level has been associated with myocardial infarction and stroke mortality (Menke et al., 2006). However, the levels were lower than \(5 \mu g/dL\), which is the number adopted by the Centers for Disease Control and Prevention as a reference value based on the blood lead level distribution in the US population (Centers for Disease Control and Prevention, 2012). Cadmium remains a determinant of cardiovascular disease mortality, even at a low level of exposure, and the average cadmium concentration \((0.15 \mu g/dL)\) was over the 80th percentile of the blood-cadmium distribution \((0.08 \mu g/dL)\) in the US population, which had attributable risks of cardiovascular disease mortality at 7.5\% (Telliez-Plaza et al., 2012). No reference value exists for the blood arsenic levels because arsenic is metabolized from the blood within a period of several hours. Therefore, urinary arsenic levels are generally considered the most reliable marker (Jomova et al., 2011). However, the average arsenic concentration \((0.82 \mu g/dL)\) was higher than the background values, which were reported to be less than \(0.1 \mu g/dL\) (Jomova et al., 2011). This average was equivalent to the blood arsenic concentrations \((0.72-1.4 \mu g/dL)\) among villagers in India who had shown DNA damage in blood lymphocytes compared to an arsenic-free group (Biswas et al., 2010). Therefore, the average concentrations of these elements were found to be not acutely toxic levels but to have attributable risks of chronic toxicity.

The concentrations of zinc, arsenic, cadmium and lead in some marine and freshwater species collected in Peninsular Malaysia exceeded their maximum permissible levels for food established by the Malaysian Food Regulations 1985 (Table S6) (Ministry of Health Malaysia, 1985). There was a positive correlation between mercury concentrations in hair and fish consumption among local communities in Malaysia (Hajeb et al., 2008; Sarmani et al., 1994). These facts suggest that the intake of local fish could be a significant source of arsenic and heavy metals in the pregnant women. In contrast, the exposure of arsenic and heavy metals from groundwater would be negligible among the pregnant women living in the capital region because groundwater is mainly used in northern states of Peninsular Malaysia but not in the capital region (Manap et al., 2013). The blood-lead levels in the present study were lower than they were when blood was collected from urban pregnant women in 1996, when levels exceeded \(10 \mu g/dL\) in 27.8\% of subjects (Hisham et al., 1998). This may reflect the fact that unleaded gasoline was introduced in Malaysia in 1992. The lead content decreased from \(0.84 \mu g/L\) in 1982 to \(0.15 \mu g/L\) in 1994, and the blood lead levels among urban pregnant women decreased from \(17.3 \mu g/dL\) in 1982 to \(7.71 \mu g/dL\) in 1996 (Hisham et al., 1998).

Nevertheless, the detection of arsenic, cadmium and lead in the pregnant women could not be associated with their contamination in the Selangor River basin because there was a limitation to directly compare the regional pollution with their blood levels in the different domain \((i.e.,\) pregnant women living in Selangor State). These could be associated if food/ground water samples in the Selangor River basin as well as blood samples of those living in the Selangor River basin were analyzed and compared their contamination levels. Because the occurrence of arsenic and lead was derived from the natural sources and the extensive land development takes place throughout the capital regions, the pollution of arsenic and heavy metals could happen in other watersheds as well, and marine and freshwater species could be significantly contaminated, as reported previously (Table S6). Thus, further investigations are necessary to identify the exposure pathways of arsenic and heavy metals to local people.

4. Conclusions

This study analyzed arsenic and 5 heavy metals in surface water and soil/ore samples in the Selangor River basin and their maternal and cord blood levels of pregnant women living in Selangor State, Malaysia. The pollution of these elements in the surface water mainly occurred at populated areas. The source proofing identified that nickel and zinc were discharged from sewage and/or industrial effluents and that lead was discharged from mining sites. The site-specific pollution of arsenic was observed in tin-tungsten deposit areas, and the pollution source could be associated with natural sources, particularly arsenopyrite. The maternal blood levels of arsenic, cadmium and lead in local pregnant women were not significantly high compared to their acute toxicity levels, but the exposure to these elements should be minimized, considering the attributable risks of chronic toxicity. They were also detected in cord blood, although cadmium and lead were significantly decreased, probably because of their prevention by amniotic membranes and placentas. Further investigations of their contamination in food and groundwater in the Selangor River basin are necessary to identify the potential exposure pathways of arsenic and heavy metals and to minimize the health risks in local people because the significant pollution of them could occur from both anthropogenic and natural sources in the capital region.

The source profiling using GIS and statistical analysis identified the pollution areas as well as potential pollution sources at a sub-basin scale in the entire watershed. This method can be applied to other watersheds because the most dataset used in GIS are publicly accessible except LandScan. The spatial analysis is helpful to squeeze the pollution areas by a sub-basin domain and to identify the potential pollution sources by combination of land use and monitoring data. Thus, this novel technique for source profiling will provide spatial information of arsenic and heavy metals pollution at a watershed scale that is useful for decision-maker to address effective countermeasures for watershed management.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.06.070.