Spatial Patterns of Heavy Metals in the Sediments of a Municipal Wastewater Treatment Pond System and Receiving Waterbody, Cha am, Thailand

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Abstract

Cha am, a popular beach destination in Thailand, uses an aerated lagoon system with four ponds in series to treat its municipal wastewater. This study investigated the spatial pattern of heavy metal concentrations in the sediment deposited at the bottom of the four ponds and along the river receiving the treated wastewater discharge. Using a stratified random sampling scheme, between 11 and 14 surface grab samples were collected from each of the four ponds on two different dates in September and October 2016 (94 samples in total). An additional 17 samples were collected in December 2016 along the 1.8 km river section connecting the ponds to the ocean. A Bruker S1 Titan 600 X-ray fluorescence (XRF) analyser was used to determine metal concentrations in the air dried sediment samples. Ordinary kriging in ArcGIS10.1 indicated that while metal concentrations were greater in the middle areas of each pond, from pond to pond the metal concentrations exhibited different spatial trends. The ponds provide treatment for most of the metals analysed, with Student *t*-tests showing that mean concentrations of arsenic, chlorine and zinc decreased significantly from the first pond to the third pond but increased significantly in the fourth pond. Chromium concentration changed insignificantly between ponds; lead concentration decreased significantly from the first to the second pond, but there were insignificant changes in mean lead concentration thereafter. Concentrations of cadmium, cobalt, mercury and selenium were below the XRF limit of detection, but the mean levels of arsenic, chromium, copper, lead and manganese in each of the four ponds frequently exceeded Ontario Ministry of the Environment and Climate Change lowest effect level (LEL) guidelines for sediment. Metal levels in the upper reach of the river, closest to the pond discharge, were similar to the pond levels and generally decreased downstream. With the exception of zinc, metal levels detected in the river sediment frequently exceeded the LEL guidelines.

1 Introduction

Because of its quiet and restive atmosphere, a 7 km long beach, and a wide range of accommodations and restaurants, Cha am, Thailand has become a popular tourist destination for both international and local Thai visitors. The population of Cha am in 2016 was reported to be 49 375. In 2001, Cha am municipality constructed an aerated lagoon (pond) system which consists of four oxidation ponds in series: an aeration pond, a settling pond, an extended aeration pond and an evaporation pond. Since tourism is an important component of the local economy and the scenic public beach is central to this tourism, it is essential to manage wastewater discharges appropriately. Past studies (e.g. But et al. 2016; Bhowmick et al. 2017; Koko et al. 2017) have examined the water quality and the efficiency of the pond system in managing

conventional wastewater parameters such as chemical oxygen demand (COD), total suspended solids (TSS), total Kjeldahl nitrogen (TKN), *E. coli* and dissolved oxygen, but to date no evaluations have assessed levels of metals in the ponds. The objectives of this study were therefore: to examine the spatial variation of metals levels, longitudinally, from the ponds on through the receiving river channel system, to the ocean; to compare metal levels in the ponds to those in the upper reach of the receiving river to assess the pond system as a source area; and to compare metal levels in the ponds and river with sediment quality guidelines to provide a first indication of environmental risk.

Although trace metals, including copper (Cu), zinc (Zn), manganese (Mn), arsenic (As), chromium (Cr) and vanadium (V) are essential components of biological functions at low levels, at higher levels metal contamination becomes a global concern

because of potential harmful effects on aquatic ecosystems and human health (e.g. Zhou et al. 2008; Khan et al. 2008; Förstner and Wittmann 2012; Tchounwou et al. 2012; Wang et al. 2013; Bjerregaard et al. 2015; Islam et al. 2015). In this study, we denote metals with a density $>5 \text{ g/cm}^3$ as *heavy metals*. Suciú et al. (2008) indicated that there are 60 such known metals in the earth–atmosphere system. Sparks (2005) noted that the term heavy metals is normally associated with issues of pollution and toxicity, even though some heavy metals are required by organisms at trace levels. Metal cycling in the environment is complex and includes emission, storage, and transformation in the atmosphere, geosphere and hydrosphere. Emissions may result from natural sources, such as biogeochemical weathering of rocks and soils or volcanic eruptions, as well as from a host of anthropogenic activities, including the combustion of fossil fuels, mining, industrial processing, transportation, industrial and municipal waste discharges, agricultural production, and urban runoff (Vermette et al. 1987; Nriagu and Pacyna 1988; Garrett 2000; Pacyna and Pacyna 2001; Irvine et al. 2005; Taylor and Owens 2009; Chen et al. 2015; Kibria et al. 2016; Nasirian et al. 2016).

It has long been known that metals preferentially adsorb to particulate matter (Förstner 1987; Förstner et al. 1990; Horowitz 1991; Gunawardana et al. 2014) and many studies have shown that up to 90% of the heavy metal loads in river systems may be particle bound (Förstner and Salomons 1980; Calmano et al. 1993; Walling et al. 2003). In a manner similar to river systems, wastewater sediment or sludge also can preferentially adsorb metals (Karvelas et al. 2003; Garcia-Delgado et al. 2007). While this propensity for adsorption can facilitate wastewater treatment through settling, the challenge of managing the subsequent sludge remains. Management options have historically included landfill, incineration, and use as an agricultural amendment due to high nutrient levels in the sludge. Concerns about the effects of metals contained in sludge on agricultural soils and crops have been raised in both developed and developing countries (McBride 2003; Jamali et al. 2009; Tiwari et al. 2011; Kumar et al. 2014; Latore et al. 2014), so it is important to monitor the metal levels in sludge when making decisions related to its use as an agricultural amendment.

Over the past 20 years, Thailand has constructed 101 municipal wastewater treatment plants throughout the country and the combined capacity of these plants is 3.2 million m^3/d (Chokewinyoo and Khanayai 2013). Among these plants, ~45% are stabilization ponds, 32% are activated sludge, 16% are aerated lagoons, 2% are constructed wetlands and 1% are rotating biological contactors. Because aerated lagoon treatment is not uncommon in Thailand, results for the case study of Cha am might be useful for other locations. Aerated lagoons, similar in design to Cha am, have been shown effective in treating conventional wastewater parameters, including nutrients, suspended sediment, COD and *E. coli*, although performance varies by design and operational

characteristics (Al-Sa'ed 2007; Li et al. 2013; But et al. 2016; Von Sperling and de Lemos Chernicharo 2017, 857). But et al. (2016) found the Cha am ponds were particularly effective in reducing TKN levels (83.2%) and *E. coli* (99.99%) between influent and effluent, but were less effective in reducing suspended solids and COD.

2 Methodology

2.1 Study Area and Data Collection

The Cha am municipal wastewater collection system is a combined system with average flows to the treatment ponds ranging between 3000 m^3/d and 5000 m^3/d during the dry season and $>10\,000 \text{ m}^3/\text{d}$ in the rainy season. The wastewater treatment ponds are located ~3 km north of the municipal area and receive wastewater discharges (as well as stormwater runoff) from the residential and commercial zones of the town (Figures 1 and 2). The aerated lagoon treatment system consists of four ponds: an aeration pond, a settling pond, an extended aeration pond, and an evaporation pond, which is part of the original wetland system in the area. Bhowmick et al. (2017) provide more detail on pond configuration and operation, but briefly the surface area and depths of each pond were: aeration pond (Pond 1): surface area 40 082 m^2 , depth 2.5 m–2.8 m; settling pond (Pond 2): surface area 16 452 m^2 , depth 2.3 m–2.5 m; extended aeration pond (Pond 3): surface area 48 522 m^2 , depth 1.7–1.8 m; and evaporation pond (Pond 4): surface area 323 612 m^2 , depth 2.0 m–2.3 m.

Cha am municipality has estimated the hydraulic retention time for the entire treatment system to be in the range of 2 weeks–3 weeks, although the water budget for the system is quite dynamic. Over a several month period in the rainy season it seems that about 59% of the water entering the ponds as runoff and direct rainfall evaporated. This percentage would increase in the dry season.

Figure 1 Thailand and location of Cha am.

Figure 2 Cha am municipal wastewater treatment pond system and flow direction.

A small river which ultimately discharges to the ocean receives the aerated lagoon effluent. The river meanders slightly and generally increases in width (from ~8 m to 30 m) and depth (from ~0.07 m to 0.4 m) as it progresses towards the ocean (Figure 3). The tidal range in this area of the Gulf of Thailand is small, with the maximum observed range in Cha am being ~2.86 m. Land use along the river is a mix of open land (some of which was formerly aquaculture ponds now infilled for development), newly constructed gated communities and townhouses set back from the river, traditional housing on the river banks, institutional (school and temple), commercial areas (restaurants, hotels, shops), and pockets of forest (including mangrove). Wastewater from the urban areas north of the river does not drain to the aerated lagoon but rather discharges directly into the river.

Figure 3 A small river connects treatment ponds to the ocean; bare land to the north is the infill from previous aquaculture ponds.

Sample locations in the ponds were determined using a stratified random approach, an example of which is shown in Figure 4. A stratified random sampling approach is frequently employed to collect environmental samples over a study area (Ferguson 1992; Caeiro et al. 2003; Ning et al. 2006; McGrew and Monroe 2009; Tóth et al. 2016). In this approach, collecting samples within the spatially-distributed grid cells helps to ensure the entire pond area is represented, but the random location of the

sample within each cell helps to preserve the unbiased structure of the data set, which is desirable in statistical analysis.

Figure 4 Stratified random sample scheme for sedimentation pond, with sample sites shown as red circles (a similar sampling scheme was used for other ponds).

Samples were collected in two different rounds, the first round on 2016-09-25–26 and the second round on 2016-10-08–09. The number of samples collected in each round for each pond is summarized in Table 1. The samples were obtained from the upper 3 cm–5 cm layer of the bed sediment using a Mighty Grab dredge device (78-080 Fieldmaster from Science First Corporation), with approximately 1 kg (wet weight) of sediment collected from each sample site.

Table 1 Number of samples taken from each pond on each sample date.

Name of Pond	Number of Samples
Aeration pond	11
Settling pond	11
Extended aeration pond	11
Evaporation pond	14
Total	47

Samples of the riverbed sediment were collected from a small boat on 2016-12-15–16. The dredge used for pond sampling was not heavy enough to sink into the river sediment and therefore was unsuitable for river sampling. Instead, sediment samples from the upper 5 cm–7 cm of the riverbed were collected by driving a length of PVC tube into the sediment and extruding the cores once they were secured safely on the boat. This approach required several cores to be collected and composited in each immediate area to obtain ~150 g (wet weight) of sediment, an amount sufficient for metals analysis. Samples were collected from a total of 17 locations along the river. Where the width of the river warranted, samples were collected on a transect at one-third, mid-channel, and two-thirds locations across the channel. Otherwise, samples were collected at mid-channel.

2.2 pH and Temperature Analysis

A portable meter (HM-20P, KK-TOA Corporation) from the Water Quality Laboratory in the Faculty of Engineering, Mahidol University was used to measure the pH and the temperature of the treatment pond sediment samples. Insufficient volume of sediment was obtained from the river sites to conduct this analysis.

2.3 Particle Size Analysis

Samples from the treatment ponds were air dried and then oven dried for 24 h at 105 °C. The samples were subsequently hand ground with porcelain mortar and pestle and dry sieved for size analysis, following Bowles (1986). The raw size data were analysed using the GRADISTAT add-in for Excel (Blott and Pye 2001). Insufficient mass was collected for the river samples to be analysed in this way.

2.4 Metal Analysis by X-Ray Fluorescence

All sediment samples (pond and river) were analysed using a handheld Bruker S1 Titan 600 X-Ray Fluorescence (XRF) system in Singapore. XRF is a non-destructive analytical method that requires no pre- or post-chemistry and delivers the concentrations of a suite of metals (much like ICP analysis) in a short run time. A large number of samples can be processed, which is ideal for geospatial analysis of contaminants. XRF analysis is not as commonly used for metals analysis as atomic absorption spectrometry (AAS) or inductively coupled plasma analysis (ICP), but it has been applied in studies as diverse as the assessment of contaminants in sediments, soils, atmospheric dust, household paints, skin whitening creams, trace element levels in alloys and geological materials, and the evaluation of art and archaeological specimens (Rhoads and Cahill 1999; Mantler and Schreiner 2000; Szökefalvi-Nagy et al. 2004; Vittiglio et al. 2004; Lawryk et al. 2009; Apeagyei et al. 2011; Diaz et al. 2014; Shuttleworth 2014; Charlou et al. 2014; Murphy et al. 2015; Lim et al. 2015; Sereyath et al. 2016; Murphy et al. 2018). Radu and Diamond (2009) showed a good correlation between XRF and AAS analysis of metals in soil samples, while Murphy et al. (2012) showed a good correlation between XRF and ICP-OES analysis of skin whitening creams.

The sediment samples were air dried thoroughly prior to analysis and any visible organic material was carefully removed. The samples were then ground with mortar and pestle. Normally about 7 g (dry weight) of sediment was placed in each XRF sample cup until full and the cup was then sealed with ultralene film. For a few samples not enough sediment was available to fill the cup and additional quality assurance–quality control (QA–QC) procedures, outlined below, were conducted to address this issue. The XRF unit was set to the soils mode library and the run time was 2 min/sample.

For QA–QC purposes certified reference material was analysed at the start of each analytical batch. The certified reference material was either the NIST SRM2710X (Montana I soils) or the CS-M2 soil from Bruker. Additional QA–QC was done by randomly selecting three pond samples and re-analysing each with four

different sample sizes (7g, 6g, 5g and 4g) to compare with the full cup results. Two other pond samples were re-analysed after a 2 d hold period in the sample cup.

2.5 Spatial Analysis of Heavy Metal Concentrations Using GIS Kriging

The geographic coordinates for each sample location were converted to a shapefile using ArcView3.3. The ArcGIS10.1 Geostatistical Analyst tool (kriging Interpolation) was then used to spatially interpolate metal concentrations based on the sample points. A variety of geospatial interpolation techniques are available in addition to kriging, including spline, natural neighbour, trend surface analysis, random forest, and support vector machine (Li and Heap 2014). Studies have indicated that kriging generally provides better results than spline and natural neighbour analysis (Voltz and Webster 1990; Laslett 1994; Pavão et al. 2012; Duong and Gourbesville 2014). Different kriging approaches have been developed (Li and Heap 2014; Forsythe et al. 2013; 2015) but ordinary kriging (OK) has been widely applied (Isaaks et al. 1989; Robertson 2008; Soffianian et al. 2015; Koko et al. 2017) and was used here for the ponds and river. The general equation for estimating a value at a point z (Chang 2012) is:

$$z_0 = \sum_{i=1}^s z_x W_x \quad (1)$$

where:

- z_0 = the estimated value,
- z_x = known value at point x ,
- W_x = the weight associated with point x , and
- s = number of sample points used in estimation.

The weights can be derived by solving a set of simultaneous equations (Equations 2, 3 and 4). For example, the following equations are needed for a point (0) to be estimated from three known points (1, 2 and 3);

$$W_1\gamma(h_{11}) + W_2\gamma(h_{12}) + W_3\gamma(h_{13}) + \lambda = \gamma(h_{10}) \quad (2)$$

$$W_1\gamma(h_{21}) + W_2\gamma(h_{22}) + W_3\gamma(h_{23}) + \lambda = \gamma(h_{20}) \quad (3)$$

$$W_1\gamma(h_{31}) + W_2\gamma(h_{32}) + W_3\gamma(h_{33}) + \lambda = \gamma(h_{30}) \quad (4)$$

and

$$W_1 + W_2 + W_3 = 1 \quad (5)$$

where:

- γ = semivariance between two known points, and
- λ = Lagrange multiplier or the weighting factor (which is added to ensure the minimum possible estimation error).

After solving the weights, Equation 1 can be used to estimate ' z_0 ':

$$z_0 = z_1 W_1 + z_2 W_2 + z_3 W_3 \quad (6)$$

3 Results and Discussion

3.1 Temperature and pH Measurements in Wastewater Treatment Ponds

July through October is the rainy season in Thailand and in September 2016 there was plentiful rainfall throughout the country. The temperature in Thailand was 0.5 °C higher than normal and the average temperature in Cha am was 28.7±0.5 °C. Thailand again was warmer and wetter than normal in October 2016, with the average temperature in Cha am being 28.1±0.4 °C (TMD 2016a; 2016b).

The minimum temperature of the sediment from the first sample date was 29.7 °C in the extended aeration pond and the maximum was 31.2 °C in the evaporation pond. The minimum value of pH from the first sample date was 6.51 in the aeration pond and the maximum was 7.4 in the extended aeration pond. The minimum temperature of the sediment from the second sample date was 29.7 °C in the extended aeration pond and the maximum was 31.4 °C in the settling pond. The minimum value of pH from the second sample date was 6.44 in the aeration pond and the maximum was 7.31 in the evaporation pond.

The sewage sludge pH from sampled wastewater treatment plants in China was reported as ranging between 4.4 and 7.7 (Wang et al. 2005), while the pH of textile dyeing sludge may range between 6.20 and 8.90 (Liang et al. 2013). Table 2 shows the correlation between the mean pH in the four ponds and the mean metal concentrations.

Table 2 Correlation between overall mean pH and metal concentrations in the sediment samples from the two sample dates.

Heavy Metal	Correlation (<i>r</i>) (pH)	
	1st Sample Date	2nd Sample Date
As	-0.7474	-0.5487
Cl	-0.7930	-0.4888
Cu	-0.8160	-0.8999
Zn	-0.8088	-0.8543
Pb	-0.7721	-0.9052
Cr	0.3982	0.6475
Mn	0.7073	0.2726

From Table 2, average concentrations of As, Cl, Cu, Zn and Pb show a negative relationship with average pH. A number of studies also have found a negative correlation between metal concentrations and pH (Perez-Esteban et al. 2014; Pereira et al. 2016; Khaledian et al. 2017). While pH is an important factor controlling the bioavailability of metals, sediment texture, aluminium and iron oxides, and organic matter (OM) also play a role (Khaledian et al. 2017). Some metals show a positive relationship with average pH such as Cr and Mn. The weakest correlation between metal and pH was for Mn on the second sample date. The environment can affect pH during the rainy season in Cha am

by impacting the minerals and microorganisms contained in the sediments. Table 2 suggests that it is important to control pond pH; should the pH become too acidic, metals can be released from the sediment.

3.2 Comparison between Overall Mean Particle Size (D_{50}) in the Sediment in Wastewater Treatment Ponds

Figure 5 compares the averaged median particle size (D_{50}) for the first and second sampling dates. Generally, the results for the two sample dates are similar except, perhaps, for the aeration pond. Sizes in Figure 5 also are within a similar range to that reported by Schmitt et al. (2011) for a wastewater treatment plant and sewer system in France (20 µm–500 µm).

Figure 5 Comparison of particle size (D_{50}) averages from the two sample dates.

The averaged median particle size (D_{50}) from both sample dates increased significantly (Student *t*-test, $\alpha = 0.05$) from the aeration pond to the evaporation pond. However, given the general characteristics of hydraulic sorting we would expect a fining from the aeration pond to the evaporation pond, which did not occur. Digestion processes in sludge or sediment influence particle size (Martinez et al. 2015). Particle size distribution will vary with field location (spatially), rainfall intensity (hydrologically), and dry season (temporally; Gulliver et al. 2010). Other factors also can affect the particle size of sediment at Cha am, such as the weather, human activity in the ponds (e.g. fishing, recreational boating in the evaporation pond), and local erosion of the soil, especially in the vicinity of the evaporation pond.

The condition of the dried sediment differed between ponds. There were no gravels contained in the sediment from the aeration pond. However, the sediment from the settling pond, extended aeration pond, and evaporation pond had increasing gravel content. The sediment from the evaporation pond also contained small pieces of shells that can affect the measurement of particle size.

Wastewater sediment is normally flocculated (Droppo et al. 1996; Liao et al. 2002; Li and Yang 2007; Yu et al. 2008) and it was expected that this would be the case for the wastewater sediment entering the aeration pond. Droppo et al. (2002) showed that street sediment generally is not flocculated, but during storm events the entrained sediment starts to flocculate in the street gutters, and flocculation occurs to an even greater extent within a combined sewer system during storm events. They also found that the larger the combined sewage floc size, the greater the settling velocity.

Flocs are a complex of organic material, extracellular polymeric substances, and discrete (smaller) mineral particles. Droppo et al. (2002) observed that when flocs were sonicated D_{50} decreased considerably, essentially because the primary particles were being measured. The unexpected trend of increasing D_{50} from aeration pond to evaporation pond may have resulted from a combination of factors, including (1) local inputs of gravel to the ponds from surrounding areas due to storm runoff and (2) the seemingly different behaviour of the evaporation pond, which is larger and part of the original natural wetland system of the area.

But et al. (2016) noted a slight increase in *E. coli* levels at the outlet of the evaporation pond (as compared to the extended aeration pond outlet) and Koko et al. (2017) reported that dissolved oxygen is consistently higher at the inlet compared to the outlet of the pond (which is opposite to the trend in the extended aeration pond). This difference in behaviour may occur due to localized runoff inputs (and is therefore linked to factor 1) and because flocculated sediment entering the aeration pond would have a greater settling velocity than the individual mineral particles that make up the floc. However, the bottom sediment size analysis was done by dry sieving in this study and this method measures the size of discrete particles, not flocs. An unexpectedly large proportion of finer mineral matter may have settled in the aeration pond due to flocculation and the sieving is therefore reflecting a higher relative composition of fine particles.

3.3 Comparison between Mean Heavy Metal Levels in the Wastewater Treatment Ponds

Mean metal levels in the sediment of each pond were calculated for both sample dates and the results are summarized in Figures 6 through 12. The figures also indicate the lowest effect level (LEL) for each metal (with the exception of Cl, for which there is no defined LEL) as determined by Ontario Ministry of Environment and Energy (MOEE 1993). LEL represents a level of contamination below which there is no effect on the majority of sediment-dwelling organisms and is therefore considered to be clean to marginally polluted.

Figure 6 Comparison of mean As concentrations in the sediment in the four ponds from the two sample dates.

Figure 7 Comparison of mean Cl concentrations in the sediment in the four ponds from the two sample dates.

Figure 8 Comparison of mean Zn concentrations in the sediment in the four ponds from two sample dates.

Figure 9 Comparison of mean Cr concentrations in the sediment in the four ponds from the two sample dates.

Figure 10 Comparison of mean Cu concentrations in the sediment in the four ponds from the two sample dates.

Figure 11 Comparison of mean Mn concentrations in the sediment in four ponds from the sample dates.

Figure 12 Comparison of mean Pb concentrations in the sediment in the four ponds from the two sample dates.

With the exception of Cl, the results for mean metals levels between the first and second sample dates are remarkably similar. The level of Cl may differ due to variable seawater intrusions into the collection system, as noted by But et al. (2016). In comparison with LEL, results in Figures 6–12 showed:

- mean As, Cr and Mn levels exceeded LEL in all ponds for both sample dates;
- mean Zn levels exceeded LEL in the aeration pond for both sample dates but were below LEL for the extended aeration and evaporation ponds;
- mean Cu levels exceeded LEL in the aeration, settling and extended aeration ponds for both sample dates but were less than LEL in the evaporation pond; and
- mean concentrations of Pb exceeded LEL in the aeration, settling and extended aeration ponds for both sample dates, but were at or below LEL in the evaporation pond.

Qualitatively, some of the mean metal levels appear to exhibit a decreasing trend from aeration pond through evaporation pond, while Cr levels seem to increase. Student t-tests were performed and the results ($\alpha = 0.05$) can be summarized as:

- As, Cl, Zn and Cu levels decreased significantly from aeration pond to settling pond for both sample dates;
- Pb levels decreased significantly from aeration pond to settling pond for the first sample date but not for the second, whereas Mn levels decreased significantly for the second sample date but not the first;
- from settling pond to extended aeration pond, Cl, Zn and Cu levels decreased significantly for both sample dates, As levels decreased significantly for the second sample date but not the first, and Mn levels showed an opposite trend, increasing significantly for both sample dates;
- Cl and Zn levels increased significantly from extended aeration pond to evaporation pond for both sample dates, while As increased significantly for the second sample date; and

- although Cr levels seemed to increase through the last three ponds, because of the high variability, this trend was not significant.

Concentrations of As, Cu, Zn, Cr and Pb for all samples were lower than the severe effect level (SEL) in each pond from both sample dates. However, Mn concentrations were higher than SEL in the aeration pond in one sample from the first sample date and three samples from the second sample date. All samples from the settling pond exhibited Mn concentrations lower than the SEL guideline, while all samples on both sample dates from the extended aeration pond had Mn concentrations higher than the SEL guideline. In the evaporation pond, five samples from the first sample date and seven samples from the second sample date exceeded the SEL guideline for Mn. There is no SEL guideline for Cl.

3.4 Correlation between Particle Size (D_{50}) and Heavy Metal Concentrations in the Sediment

Pearson product moment correlation analysis was used to analyse correlations between sediment D_{50} and metals levels. The results are shown in Table 3. D_{50} was significantly correlated ($\alpha = 0.05$) with levels of As, Cu, Zn and Pb but not Mn, Cl or Cr. Although desirable, we did not directly analyse the metals levels in different sediment size classes. Studies have frequently shown finer sediment sizes are associated with higher metal concentrations, in part as a result of clay mineralogy and adsorption (e.g. Förstner and Salomons 1980; Singh et al. 1999; Murray et al. 1999; Maslennikova et al. 2012) and the negative correlation results in our study generally are consistent with this size trend.

Table 3 Correlation between the overall mean particle size (D_{50}) and metals concentrations in the sediment samples from the two sample dates.

Heavy Metal	Correlation (r) (Particle Size)	
	1st Sample Date	2nd Sample Date
As	-0.8330	-0.5569
Cl	-0.5516	-0.4966
Cu	-0.9005	-0.9002
Zn	-0.8700	-0.8555
Pb	-0.7948	-0.9040
Cr	0.3727	0.6480
Mn	0.2149	0.2861

3.5 GIS Ordinary Kriging Interpolation: Treatment Ponds

Ordinary kriging (OK) was used to assess spatial patterns of metals in each pond. Unlike the kriging results reported by Koko et al. (2017) for the ponds where there were regular spatial patterns for dissolved oxygen, it is more difficult to generalize about the spatial patterns for metals in the sediment. The spatial patterns of Cu and Pb concentrations in the sediments from the aeration pond are shown in Figures 13 and 14.

Figure 13 Spatial pattern of Cu in the aeration pond (upper: first sample; lower: second sample).

Figure 14 Spatial pattern of Pb in the aeration pond (upper: first sample; lower: second sample).

Figure 15 Spatial pattern of Cl in the evaporation pond (upper: first sample; lower: second sample).

Figure 16 Spatial pattern of Mn in the evaporation pond (upper: first sample; lower: second sample).

The Cu patterns show that concentrations in the sediment were higher in the middle and the east side near the influent of the aeration pond for both sample dates (although the trend is stronger for the first sample date). The second sample date also exhibited a distinct zone of lower Cu concentration near the effluent point. Lead patterns show that concentrations were higher on the east side of the aeration pond for both sample dates. The highest concentrations of Pb in the sediment were near the influent for both sample dates.

In the evaporation pond, metal concentrations can exhibit strong but opposite spatial trends, for example, Cl and Mn (Figures 15 and 16). Cl patterns from the two sample dates were higher near the influent of the evaporation pond, while Mn concentrations were higher near the effluent of the evaporation pond.

3.6 Trends in Metal Levels for the Receiving Waterbody

The metals levels in the bed sediment of the small river receiving the treated discharge from the pond system are summarized in Table 4 and Figure 17. The data in Table 4 show that, with the exception of Zn, metals levels in the river sediment frequently exceeded LEL, while one site exceeded the SEL for Mn and two sites exceeded the SEL for Cu.

Table 4 Metals levels in bed sediment of receiving waterbody (small river), mg/kg.

Sample	Cl	Cr	Mn	Cu	Zn	As	Pb
1	14 437	36	551	24	98	15	38
2	5144	55	472	63	26	10	21
3	5819	70	355	39	78	7	35
4	11 952	28	588	127	85	15	34
5	9311	53	472	18	59	11	29
6	10 710	10	371	53	48	1.5	42
7	12 220	57	505	55	44	7	28
8	17 586	47	949	12	43	12	5.5
9	22 488	72	783	35	77	7	25
10	16 896	32	615	21	23	2	28
11	11 370	105	389	18	1.5	1.5	5.5
A1	10 551	31	481	37	124	14	55
A2	15 957	100	180	8	7	1.5	23
B1	15 151	40	760	34	80	7	5.5
B2	9456	47	1211	11	29	6	25
C1	4708	46	461	44	43	10	28
C2	13 212	23	692	147	134	10	58
MOEE LEL	NS	26	460	16	120	6	31
MOEE SEL	NS	110	1100	110	820	33	250

The ordinary kriging in Figure 17 indicates that levels of As, Pb, and Zn generally decrease downstream from near the evaporation pond to the river mouth. Chromium levels also decrease from near the evaporation pond to approximately 0.9 km downstream, but then increase, with the highest levels located near the river mouth. Copper decreases from near the evaporation pond

towards the mouth of the river, but there is an unusual hotspot near the aquaculture ponds that have been infilled. The levels of Mn show no particular spatial pattern along the river and are quite similar overall.

Figure 17 Ordinary kriging of metals levels in the receiving river downstream of the treatment ponds (top-bottom: As, Pb, Cu, Cr, Zn, Mn).

Table 5 compares the metals levels in the river and the evaporation pond to explore whether the pond has an impact on the receiving water.

Table 5 Metals levels in evaporation pond and upstream river sediment.

Metal	Evaporation Pond Concentration, Second Sample	Concentration, upstream-most river
	Date, mg/kg	site, mg/kg
Cl	2193	14 437
Cr	43	36
Mn	1077	551
Cu	15	24
Zn	73	98
As	3.72	15
Pb	30	38

Results in Table 5 must be interpreted cautiously because of three factors. First, while the evaporation pond is at the downstream end of the treatment train, as is noted above, the upstream ponds also have a role in reducing metals levels, which is not considered in the comparison between the evaporation pond and the river.

Second, the river may be impacted by localized metal inputs. As noted, aquaculture ponds were previously located in an area near the head of the river, but these have been infilled for development. The fill consists of loose sand and some of this sand has migrated locally into the river channel. The sand might serve to

reduce metals levels in the riverbed, although in the case of Cu the sand may be a local metal source and produce a hotspot. Unfortunately, the source of the sand is unknown. Levels of Cr appear to increase along the lower half of the river, with the highest concentrations near the mouth. Figure 17 shows that the area of increasing Cr coincides with the more dense urban and commercial land uses near the waterfront. It is possible that anthropogenic activities and stormwater runoff from these areas is a local source of Cr, as higher vehicle activity (brake and tire wear), yellow street paint, and corrosion of stainless steel restaurant appliances might be sources of Cr (Adachi and Tainosho 2004; Apeagyei et al. 2011; Kaladhar et al. 2012; Tang et al. 2013).

Third, because the river discharges to the Gulf of Thailand, it will be influenced by tidal cycles and backwater effects. The tidal range in this area of Thailand is not large and some preliminary river model results using PCSWMM suggest that tidal influence is limited to the lower half of the river. However, to date we have inconsistent results from sampling the conductivity of the river water. In July 2016, conductivity was relatively low in the upper river, in the range of 2 mS/cm–4 mS/cm (consistent with PCSWMM results), but in December 2016 conductivity in the upper river was 32 mS/cm–35 mS/cm, which approaches levels found in seawater. The higher Cl level in the sediment for the upstream river site (Table 5) may reflect greater tidal influence during December 2016 but these dynamics need to be investigated further. Levels for the other metals in the evaporation pond and upstream river area are comparable, so this system seems to completely treat the inflow by the end of the evaporation pond. The general trend of As, Pb and Zn concentrations decreasing in the downstream direction is consistent with transportation from the evaporation pond source area. The localized inputs of Cr, in particular, could be influenced by tidal movements near the river mouth.

3.7 QA–QC Analysis

Certified reference material was run at the start of each sample batch as a QA–QC measure for the Bruker Titan S1 600 XRF unit. For the pond sediment, the Bruker CS-M2 standard was used, while for the river sediment NIST 2710a Montana I soil was used. Table 6 shows the results of QA–QC with the Bruker CS-M2 standard and Table 7 shows the results for the NIST reference material.

Table 6 QA–QC analysis for XRF with Bruker CS-M2 standard.

	Metal Concentrations (mg/kg)				
	As	Zn	Cr	Cu	Mn
Bruker CS-M2 standard	76	713	66	194	991
QA–QC for the first sample date	98	769	72	204	997
QA–QC for the second sample date	76	759	65	198	983

Table 7 QA–QC results for NIST 2710a Montana I soil.

	Metal Concentrations (mg/kg)						
	Cl	Cr	Mn	Cu	Zn	As	Pb
NIST Standard	<LOD	23	2140	3420	4180	1540	5520
Bruker	<LOD	31	2023	3261	4223	1532	5692

LOD: Limit of Detection; for Cl LOD was 135 mg/kg.

The metals levels determined for both reference materials were quite similar to the CS-M2 and NIST 2710a Montana Soils I certified reference material. Additional QA–QC was undertaken by randomly selecting five treatment pond samples from the first sample date. Three of the five selected samples were analysed with four different sample sizes (weight) and the mean values were compared with the results obtained for the corresponding samples analysed previously, to determine if the air in the sample cup affected the measurement of the metals concentrations. Tables 8 and 9 show the results of the additional QA–QC.

Table 8 Comparison of metal concentrations in five selected sediment samples with the corresponding samples analysed earlier.

Sample (random)	Metals Concentrations (mg/kg)					
	As	Cl	Zn	Cr	Cu	Mn
A1 136 (7 gr)	18	4109	417	30	78	983
A1 136 (6 gr)	18	4309	403	25	81	950
A1 136 (5 gr)	28	4344	391	14	81	1016
A1 136 (4 gr)	20	4295	427	31	98	993
A1 136 (earlier sample)	18	4236	416	41	81	966
A2 158 (7 gr)	11	3094	130	24	35	890
A2 158 (6 gr)	10	3030	142	24	31	937
A2 158 (5 gr)	14	3059	123	9	29	920
A2 158 (4 gr)	12	3180	126	34	30	847
A2 158 (earlier sample)	10	3080	131	24	34	878
A3 163 (7 gr)	4	1013	49	49	15	2392
A3 163 (6 gr)	8	1028	50	69	19	2378
A3 163 (5 gr)	3	868	46	54	14	2405
A3 163 (4 gr)	10	842	45	41	21	2241
A3 163 (earlier sample)	10	1034	44	51	14	2422
A3 171	13	2154	55	24	20	3077
A3 171 (earlier sample)	11	2150	51	15	20	2923
A4 185	17	3187	76	35	17	995
A4 185 (earlier sample)	15	3336	71	30	16	1054

Table 9 Comparison of metals (expressed as a percentage difference of the earlier sample) in the five selected sediment samples with different masses.

Sample (Random)	QA–QC after Analyzing the Samples (%)					
	As	Cl	Zn	Cr	Cu	Mn
A1 136 (7 gr)	100	97	100	73	96	102
A1 136 (6 gr)	100	102	97	61	100	98
A1 136 (5 gr)	156	103	94	34	100	105
A1 136 (4 gr)	111	101	103	76	121	103
A2 158 (7 gr)	110	100	99	100	103	101
A2 158 (6 gr)	100	98	108	100	91	107
A2 158 (5 gr)	140	99	94	38	85	105
A2 158 (4 gr)	120	103	96	142	88	96
A3 163 (7 gr)	40	98	111	96	107	99
A3 163 (6 gr)	80	99	114	135	136	98
A3 163 (5 gr)	30	84	105	106	100	99
A3 163 (4 gr)	100	81	102	80	150	93
A3 171	118	100	108	160	100	105
A4 185	113	96	107	117	106	94

The percentages in Table 9 show the differences between the metals concentrations in different sample sizes and the metals concentrations obtained for the corresponding earlier samples. The results show that most of the differences were not large between the five selected samples with four different sample sizes (weight) and the corresponding samples analysed earlier.

4 Conclusions

With the exception of Cr, metals levels in the sediment of the aerated lagoon system tended to decrease moving from the head of the system towards the outlet. The sediment in the ponds, on average, was lightly contaminated by all metals (i.e. exceeded LEL), although Cu and Pb, on average, were less than LEL in the evaporation pond. Only individual samples of Mn exceeded the SEL guideline. Ordinary kriging identified spatial trends in metals levels that might be considered should the sediment be dredged and recycled for agricultural purposes. For example, Pb levels were higher on the eastern side of the aeration pond than in other parts of the pond and such areas with elevated levels need to be handled with greater care.

With the exception of Zn, metals levels in the river sediment frequently exceeded LEL, while one site exceeded SEL for Mn and two sites exceeded SEL for Cu. Metals levels in the upper reach of the river tended to be similar to those observed in the evaporation pond and decreased downstream towards the Gulf of Thailand. This trend is consistent with the treatment ponds being the source area. There are some local hotspots for metals and Cr, in particular, showed a reverse trend to the downstream decrease in metal levels. The levels of Cr were greatest near the mouth of the river and likely reflect local inputs from the more dense residential and commercial area near the waterfront.

The handheld XRF unit is a useful tool to rapidly provide accurate levels of total metals in sediment. This is particularly attractive, in combination with geospatial visualization techniques, for identifying hotspot areas and supporting sediment management decisions. It should be noted, however, that XRF analysis provides data only on total metal levels, and is therefore a less powerful indicator of potential toxicity and adverse health effects. To address this shortcoming, some type of sequential extraction should be conducted on the sediment to assess the relative mobility of particle-associated metals (Amir et al. 2005; Lee et al. 2005; Irvine et al. 2009; Nemati et al. 2011; Liang et al. 2013).

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