ENVIRONMENTAL ENGINEERING
ASSOCIATION OF THAILAND

THAI ENVIRONMENTAL ENGINEERING JOURNAL

Vol. 34 No. 3 September – December 2020 ISSN 1686 - 2961
ISSN 2673 - 0359 (Online)

Thai Environmental Engineering Journal

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Thai Environmental Engineering Journal

Vol. 34 No. 3 September – December 2020

ISSN 1686 - 2961
ISSN 2673 - 0359 (Online)

Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 1-9
www.eeat.or.th

Investigation of Contaminated Soil Formed at River Bank Located Downstream of Mines

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Abstract

The purpose of this study is to find out how polluted a riverbank located downstream of the mines where has been excavated for more than 100 years in Serbia. A field survey was conducted, and ground surface sediments from rivers were collected, sediment thickness and density were measured, and soil pits were excavated. The inside of those pits were observed and the sediment samples were collected. The surface sediments collected were subjected to mineral identification using XRD, and the sediments collected from the pits were subjected to chemical analysis. On the other hand, a high-accuracy ortho-image was created by using UAV to acquire visible images with 60% or more of each other overlapping and by acquiring position information using high-accuracy GPS. From this image, the area of the riverbank was measured using GIS software. In addition, a camera capable of acquiring hyperspectral images was installed in the UAV to acquire hyperspectral images. From the obtained image analysis results, it became clear that pollutants from the mine were distributed on most of the surface of this riverbank. The conclusion of this study is that there is a high possibility that the waste material from the mine had been accumulated at the riverbank that was the subject of this study, and the area was occupied with 96,000 m^2, the volume is 78,600 m^3 in volume and 152,400 t in weight.

Keywords : contaminated soil; heavy metal; field survey; UAV; jarosite

Introduction

Contaminated soil is produced in mines and under geological conditions. Especially in the vicinity of the mine, the pollutants are suddenly discharged and diffused from the start of excavation, which causes serious environmental pollution. The polluted soil discharged by mining moves on the ground surface by the force of water and wind like other debris. It is known that contaminated soil carried by river water behaves similarly to other debris [1]. However, without knowing the actual status of the deposited pollutants, it is not possible to discuss environmental pollution status, future diffusion potential, environmental impact, and measures to improve them. Therefore, in this study, we will report the results of field surveys and environmental surveys using UAVs for the current state of sediments in rivers that have been transported and deposited by river water around the mine.

Study Area

Bor and its surroundings in eastern Serbia are well known for copper deposits, which are among the largest in Europe (**Figure 1**). Around these deposits, many current and historical copper mines were developed since 1903. Our study area is the floodplain of a river located downstream of the Bor mine. The Bor mine has been mined upstream of the Bor and Krivelj rivers and started in 1903 and 1979, respectively. They are called Old-Bor mine and Veliki Krivelj mine (**Figure 1b**). The study area is a riverbank formed at the confluence of these Bor and Krivelj rivers (**Figure 2a, c**).

Underground mining started at Old-Bor mine in 1903 and open pit excavation began in 1923. Drilling was continued until 1993. During that period, mine waste was directly discharged into rivers until 1933, after that, overburden, low-grade ore, and tailing were deposited around the mine. At Veliki Krivelj mine, excavation of the open pit started in 1979, and overburden, low grade ore and tailing are deposited around the open pit. Direct disposal to the river at the beginning of excavation at Old-Bor mine is a problem. Regarding the tailings, the dam constructed as a sedimentation site is the dam that built up the tailings themselves, and is not fixed or covered (**Figure 2b**). It is not difficult to imagine how these will behave due to rainwater or river runoff.

In the study area, rivers with debris are deposited on the Bor river and Krivelj river in front of the confluence, and after the confluence, a wider riverbank is formed (**Figure 2**). Figure 2a shows the riverbank formed along the Bor river, and Figure 2c shows the confluence of the Bor and Krivelj rivers.

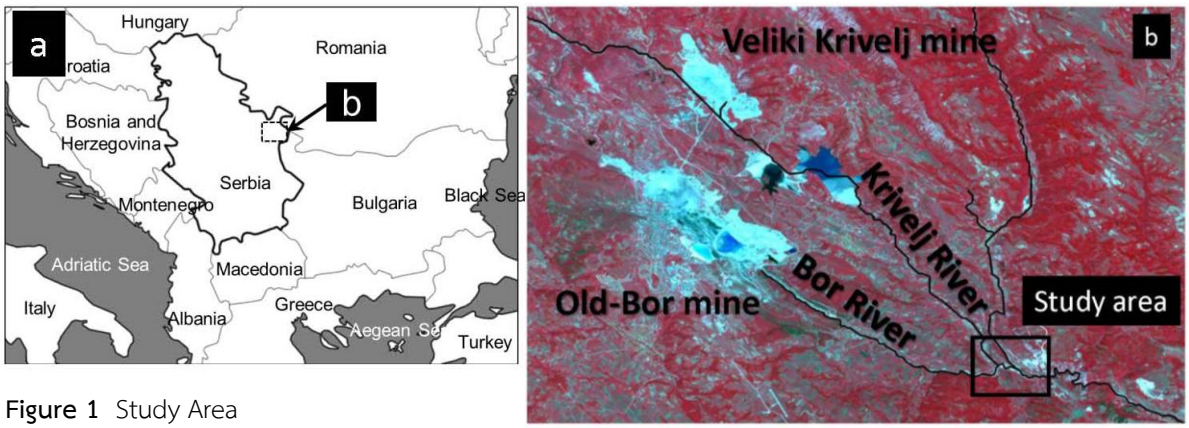

Figure 1 Study Area

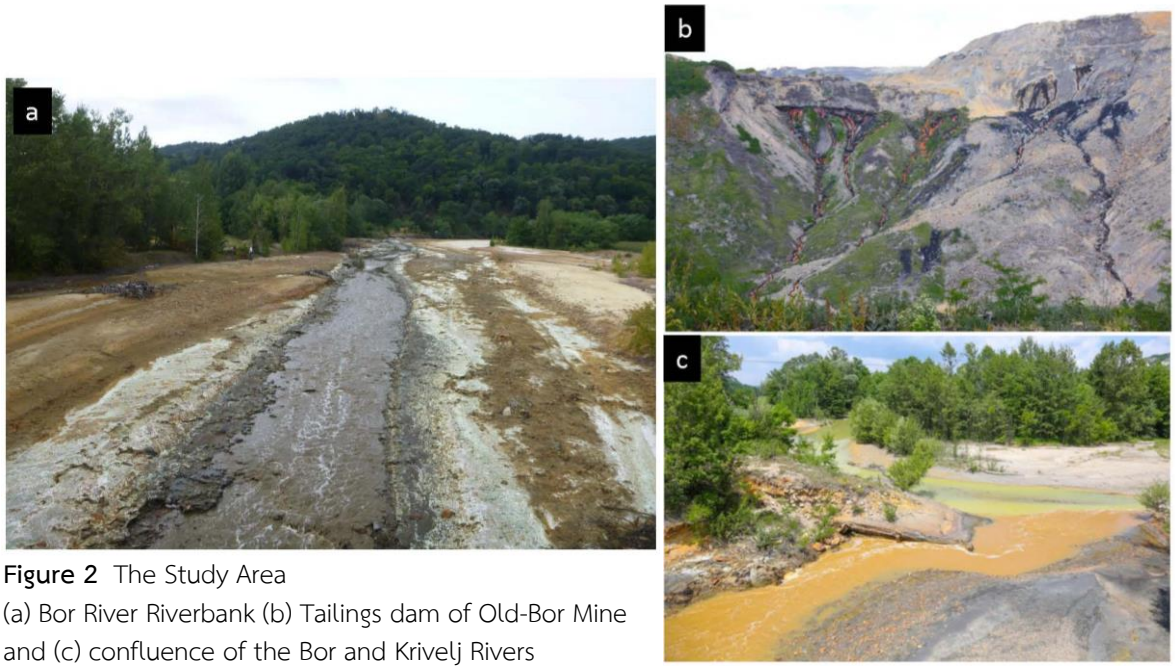

Figure 2 The Study Area
(a) Bor River Riverbank (b) Tailings dam of Old-Bor Mine and (c) confluence of the Bor and Krivelj Rivers

Methodology

To clarify the current condition of the target river basin, we conducted a field survey, analyzed the collected samples in the laboratory, and analyzed the images acquired by UAV.

In the field survey, surface material was sampled, then the mineral composition of the samples was investigated using X-ray diffraction analysis (XRD). Riverbanks sediments developed along each river were excavated, and sedimentary facies of the pits were observed. The sediment samples were also collected from the excavated pits. The sediment samples were analyzed for chemical content by ICP-MS and FAAS. The depths of sediments were investigated using a cone penetration test. The density was obtained by measuring the weight of the ground surface sample collected in a fixed form on the spot.

Two types of images were acquired using UAV. One is a set of visible images in which adjacent places overlap to create a high-precision digital topographic map. The set was taken using GoPro camera mounted on Phamtom3. The other set is hyperspectral images for classifying the reflection properties of surface materials. A total of 46 hyperspectral images were acquired every 10 nm in the range of 600-1050 nm. Since it was necessary to change the exposure time during shooting, the shouting was carried out twice and the images were gotten a total of 92. An LCTF camera was used for the image acquisition [2]. The aerial photography carried out with the LCTF camera mounted on the UAV was carried out by requesting a specialized UAV pilot on 6th September, 2018.

To create a high-precision topographic map, the acquired photographs are made into point clouds using photo composition software (PhotoScan, Agisoft). The position information was also acquired by UAV, but it was corrected by the ground control point (GCP) acquired by high-precision GPS locally (Trimble R4). After correction, the area of the riverbank was

measured from the created ortho image using ArcGIS software (ESRI).

The photographs taken by the LCTF camera were corrected geometrically for 46 images of each set, then normalized using grey mat corrected in laboratory. Finally, the two set were georeferenced and merged. From the merged image, a spectral curve can be obtained for each pixel. However, it was difficult to correlate with the spectrum curve obtained by the existing spectrometer. On the other hands, there was a difference in the spectrum for each pixel, and it was possible to perform spatial analysis on the same image. Therefore, supervised data was acquired from the obtained image, and supervised analyses was performed (Here, the SAM method was adopted).

Results and Discussions

XRD of surface samples revealed that it contains jarosite, which is often found around mines where pyrite is often produced. Therefore, as shown in **Figure 3**, the quantity ratio of jarosite at the sampling point is shown by circles: large content: L for red, medium content: M for green, and small content: S for blue. Jarosite is a secondary mineral of iron oxide minerals and is a recrystallized product of mine waste dissolved in water [3]. In addition, since water used for crystallization is around pH2 [3], it indicates that acidic water is present in the environment where jarosite is produced.

Soil pits were dug on the left bank of the Bor and the right bank of the Krivelj. **Figure 4** shows each of the soil faces and the concentrations of copper, lead, arsenic, and manganese measured by ICP-MS and iron and sulfur concentrations measured by FAAS for each sedimentary structure. There are six sedimentary layers in the pit of the Bor river, and alluvial deposits were observed beneath them. The concentration of alluvium is lowest. The concentration of the second layer of the Bor river bank is missing and not shown. In the sixth layer at a depth of 62-120 cm, sub-angular gravels with a maximum diameter of 20 cm were observed, and the concentrations of copper, iron and sulfur were high at 1,900 ppm, 17%, and 27% respectively. Lead and arsenic had the highest concentrations in the uppermost layer, 220 ppm and 240 ppm, respectively. Manganese concentration was highest in the second layer. In the uppermost layer, copper and iron were also generally high in concentration and were 550 ppm and 4%, respectively.

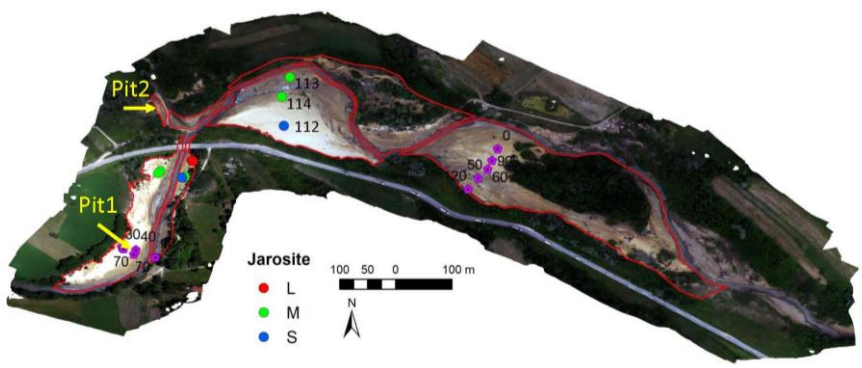

Figure 3 Field survey data on the orthoimages created in this study

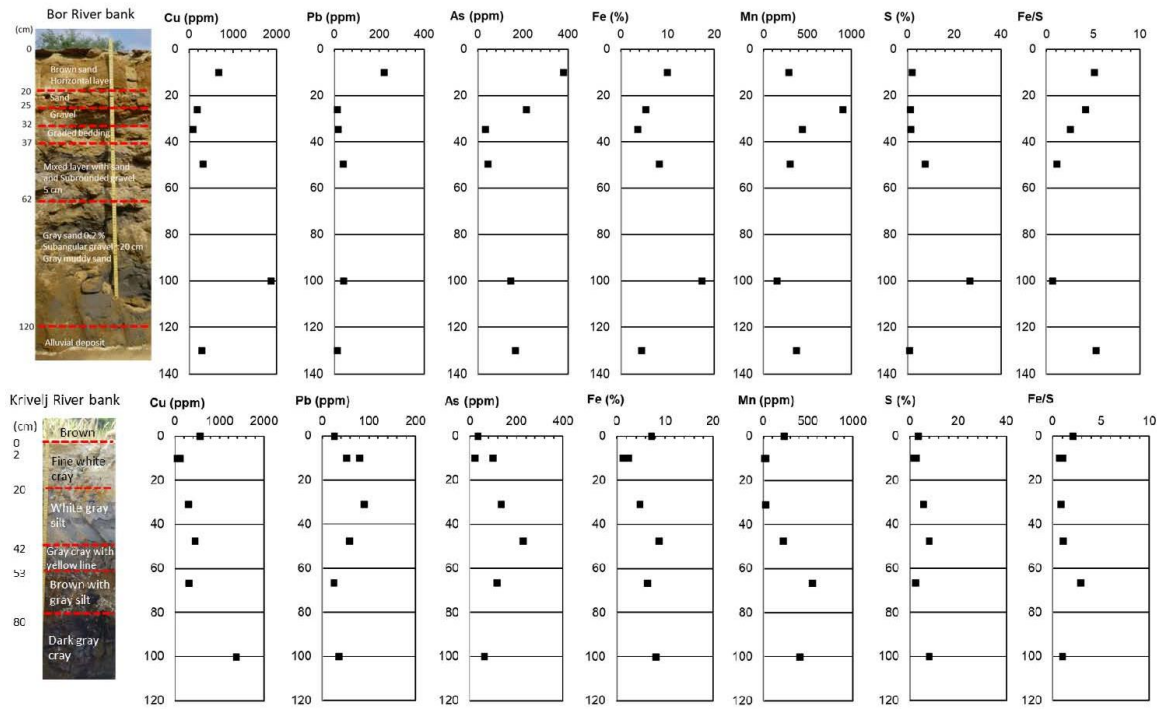

Figure 4 Soil faces and concentrations of copper, lead, arsenic, and iron of each layer of pits

As mentioned above, at Old-Bor mine, for about 30 years after the start of mining in 1903, mine waste was directly discharged to rivers. The 20 cm-sized gravel found at the sixth layer of the Bor riverbank deposit is considered to be waste at that time. This can also be explained by the extremely high concentration of copper, iron and sulfur in the bottom layer. It can be estimated from the mining record of the Old-Bor mine that the copper concentration was 6% for the first 30 years [4]. It is presumed that the gravel with such a large diameter had flowed down since the early 1933, as waste was deposited on dams and sedimentation sites. On the other hand, in the upper deposit, the grain size of sand to silt is accumulated in layers, and the grade structure is seen in the fourth layer. It can be inferred that these deposits flowed down after the tailing dam was formed and when the dam wall collapsed due to a flood. The depth profiles of the concentrations except manganese and sulfur are similar. It can be inferred that this sediment does not move much physically from the time of deposition and represents the situation at the time of deposition. Manganese and sulfur are easier to move than iron. Especially in the 1st to 3rd layers, the amount of sulfur is relatively small, which suggests that the upper layer is being oxidized. Further, the fourth and fifth layers had a high sulfur concentration, and their ratios to iron were 1.1 and 0.7. In other words, the area around this depth is in an anaerobic state, indicating that the oxidation of pyrite has not progressed. Probably indicates that the area around this layer is the water table.

Six sedimentary layers were also observed in the Krivelj river pit, but the structure of the sediments was very different from that of the Bor river. In addition, due to the water table, it was not possible to dig into a layer below the bottom layer. The pit deposits on the Krivelj River were fine, mostly clay to silt size. Strong elasticity and plasticity were observed in the

dark gray clay layer at the bottom. Even in the Krivelj River pit, the concentration of copper in the bottom layer was the highest, at 1,400 ppm. On the other hand, the concentrations of iron and arsenic were high in the middle white gray silt layer, 8.9% and 230 ppm, respectively. Manganese and sulfur concentrations did not differ from those found in Bor. The Fe/S ratio was 1 or less than 1 in most layers. Only in the layer of 53 to 80 cm, the Fe / S ratio was 2.9. The color of this layer is brown, consistent with the oxidation and sulfur elution that would have occurred.

The sediments of the Krivelj riverbank are even finer. This is convincing evidence that the tailings produced during excavation and beneficiation after 1979, when technological innovation has passed, may have flowed down. The investigation of these particle sizes was only visual and could not be accurately measured. The difference in concentration in each layer may be interpreted by comparing it with the drilling record. However, the behavior of heavy metals in soil must also be considered, and this study has not considered it. I would like to make it a future issue. Both Bor riverbank and Krivelj riverbank have six sedimentary layers. In order to interpret this, it is necessary to carry out more detailed topographical survey. On the other hand, it was estimated that the Bor riverbank deposits were brown and had less sulfur than when they were deposited. It can be estimated that sufficient time has elapsed for oxidation. On the other hand, in the Krivelj riverbank, white and gray deposits are present, suggesting that they have not yet been oxidized. Bor deposits should be from over 100 years old and Krivelj deposits from the last 40 years. What is the reason for this difference in deposition period and the same number of layers? There are several possible reasons for this. (1) During the first 70 years, deposits from Old-Bor mine were deposited on the Bor river bank. As a result, the riverbed of the Krivelj river was lowered, and the environment became prone to deposits in the Krivelj riverbank. (2) Since the Krivelj River has a larger catchment area than one of Bor river, sediments are more likely to collect. (3) Veliki Krivelj mine has undergone technological innovation and has undergone large-scale construction, so the amount of sediment has increased rapidly, and a large amount of sediment has come to flow suddenly. (4) Lots of bedding is visible in the sediments of the Bor riverbenk, and there may be other bedding that cannot be distinguished in this observation. In any case, since all the layers contain chemical substances that are considered to be of mine origin, it can be inferred that all of these deposits were mine waste.

As observed in the two soil pits, the deposit thickness was 120 cm and deeper than 100 cm, respectively. In addition, the thickness of sediments in the riverbank was examined by a portable corn penetration tester. The survey points are the purple hexagonal points shown in **Figure 3**, and the results are the numbers written at each point. The depth near the river channel was deep, and the one far was shallow. The average of the Bor riverbank was 70 cm, and the one of the Krivelj and after junction riverbank was 84 cm. Overall, the average sediment depth was 76 cm. On the other hand, the density of surface sediments was also measured in the riverbank of Bor river, Krivelj river, and after the junction. Their values were 1.76 g/cm^3, 1.76 g/cm^3, and 1.97 g/cm^3, respectively.

Figure 3 shows the ortho image created from the visible images. This image was position corrected with 15 GCP points measured by precision GPS sensor. In this study, the area of riverbank sediment was estimated from this ortho image. Polygons were created with the area surrounded by the red line in the figure as riverbank sediment, and the area of the polygon was calculated. As a result, the total area of riverbank sediments in this area was estimated to be 96,000 m^2.

The volume of riverbank sediment was calculated based on the sediment density, the average depth and area of the sediment from the field surveys in this study. The volume of the riverbank was calculated simply by dividing it into the Bor riverbank, the Krivelj riverbank, and the riverbank after confluence. The result was calculated as 18,400 t, 2,600 t and 131,400 t, respectively. Therefore, the total was 152,400 t. The volume was 78,600 m^3. In other words, if it is necessary to cover contaminated soil for environmental protection, it is necessary to work to cover the ground surface of 96,000 m^2 or more. Also, if contaminated soil is to be removed, 152,400 t of soil must be transported and deposited.

Obtained 92 hyperspectral images was position and color corrected. As the result, spectral data of 600 to 1050 nm was obtained for each pixel. The obtained spectrum data from the images and the ground truth samples are shown in the **Figure 5**. For the ground truth samples, it was assumed that the stronger the jarosite peak intensity during XRD, the higher the amount of jarosite. The medium amount is Jarosite M, and the small amount is Jarosite S. The spectral curves of the ground truth samples in Figure. 5 were the result of analyzing them in the laboratory using portable spectroradiometer (FieldSpec). The reference values for jarosite were obtained from the USGS spectral library [5], and the values were normalized and shown in **Figure 5**. Among the reflection characteristics of jarosite, the absorption band seen near 900 nm is one of its characteristics [6]. The spectral characteristics of the ground truth sample show which absorption bands are slightly visible. The spectrum curve obtained from the hyperspectral image is not clear, although it can be seen depending on the appearance. It is necessary to conduct more statistical analysis in the future, but in this study, we will limit the interpretation to this point. Therefore, the spatial distribution analysis of the by selecting the supervised data from the own image (**Figure 6**). The supervised data was selected by field survey to determine the ROI range of trees, roads and rivers. For Jarosite M and Jarosite S, ROI was created around the point where each ground truth sample was collected (area surrounded by squares in **Figure 6**). Spectral Angle Mapper (SAM) method was used for supervised classification.

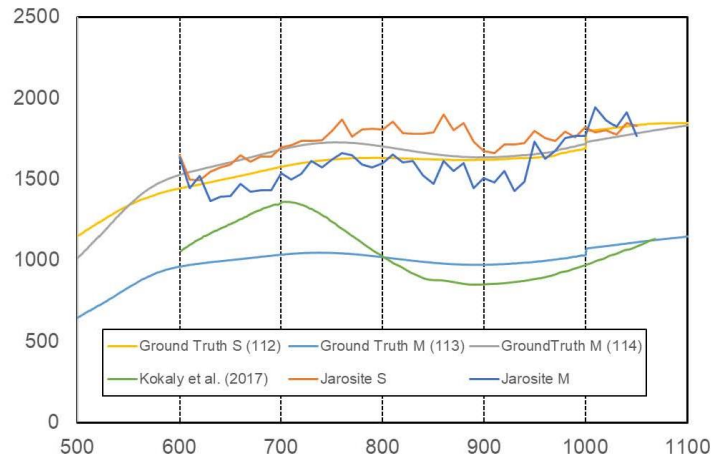

Figure 5 Spectral features obtained from hyperspectral image and ground truth samples

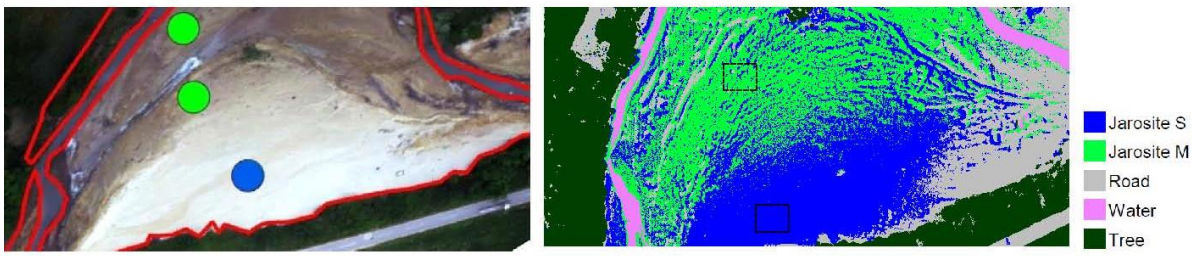

Figure 6 Spatial classification analysis result. The left side is the relevant part of the analysis in Figure 3

Spatial distribution analysis results show that components with the same spectral characteristics as roads also appeared on the surface of the sediment, and it is hard to say that it was completely successful. However, the classification of trees and water is well done, and the classification of Jarosite S and Jarosite M brings interesting results. It is regrettable that many roads have been included, but the control ratio in the Figure 6 for each component is shown in **Table 1**. From the distribution characteristics of Jarosite M and Jarosite S, the distribution of Jarosite M tends to be closer to the river. This may suggest that the frequency of inundation affects the oxidation of surface materials. Also, it seems that Jarosite M covers Jarosite S. This might indicate that the more recently deposited material contained more M, or that the sheet flood flow made the surface material more oxidized. Furthermore, the planar structure of the deposit can also be estimated from this figure. In the area close to the river channel, a lump-like, band-like, and sedimentary structure along the flow channel is seen, whereas in the area far from the river channel, it seems to have a granular and individual distribution. On the other hand, what does the surface sediment classified as the road represent? Usually, road materials are used in the surrounding area, so it is not so strange that the same materials come out. Also, in Figure. 6, it seems to be distributed far from the river channel, further below the jarosites M and S, and at the most downstream of the meandering part of the river. This may be due to the fact that sediment oxidation has not occurred in this area or the acid stream flow has not reached this far. In any case, the result of this analysis of hyperspectral images is that the sediment containing jarosite covers most of the riverbank. Whether this is changing over time will be answered by reacquiring the image at a later time. Since it is a UAV-based research, it may be a relatively easy future, but I look forward to future research.

Table 1 Distribution ratio of each component in Fig. 6

Image Class	Samples	Percent
Jarosite S	56,132	32.0
Jarosite M	48,945	27.9
Road	31,130	17.7
Water	5,371	3.10
Tree	33,826	19.3
Total	**175,404**	**100**

Conclusion

In this study, we used geoscience techniques and UAV techniques to investigate the state of the sediment in a riverbank located downstream of a mine. It was found that the target deposit contained jarosite, a secondary iron oxide mineral. In addition, inspection of the sediment structure of the two soil pits and chemical analysis of the sediment samples confirmed the presence of mine waste deposits in the riverbank. Besides, we estimated the area and amount of contaminated sediment containing these, and obtained the necessary numbers to remove these contaminations. In addition, the hyperspectral image obtained by UAV confirmed that the pollutant was spreading over the entire ground surface.

Acknowledgement

This research was partially supported by the Japan Science and Technology Agency (JST) and the Japan International Cooperation Agency (JICA) through Science and Technology Research Partnership for Sustainable Development (SATREPS). This work was also supported by JSPS KAKENHI Grant Number 17K17613.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 11-21
www.eeat.or.th

Composition and Functional Responses of Microbial Community to Temperature and Substrate in Anaerobic Digestion Process

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Abstract

This study investigated the impact of two important key factors including temperature and substrate on microbial community in the anaerobic co-digestion process of septage and longan peel waste by temperature phased anaerobic digesters (TPAD). Denaturing gradient gel electrophoresis (DGGE) and metagenomics sequencing was used to analyzed microbial community structures. The DGGE and cluster analysis results clearly indicated that substrate and temperature strongly influence the structure of bacterial populations. Significant differences of microbial communities were observed from both TPADs digesters. Also, co digestion with longan associated with the changes of bacterial community structure in TPAD system. It was found that *Firmicutes Bacteroidetes Cloacimonetes Tenericutes and Proteobacteria* were most dominant bacterial phyla in TPAD systems. High number of *Firmicutes* and *Tenericutes* were detected from mesophilic tank, while *Bacteroidetes* and *Cloacimonetes* were found from thermophilic reactor. Moreover, each of the digesters harbored distinct yet dynamic microbial populations, and some of the methanogens were significantly correlated with methane productions. *Methanosarcina* and *Methanothermobacter* appeared to be the most dominant methanogenic genera in both digesters operated with different temperatures. The microbiological findings may help understand the metabolism that underpins the anaerobic processes within each of the two digesters of TPAD systems co-digesting septage and agricultural waste.

Keywords : Anaerobic digester; Temperature Phased Anaerobic Digestion (TPAD);
PCR-DGGE technique; Microbial community

Introduction

Anaerobic digestion and its designs including Temperature Phased Anaerobic Digestion (TPAD) have been developed and applied for various types of waste and wastewater including agricultural waste for a source of renewable energy [1-5]. In recent years, the implementation of anaerobic co-digestion has gained a lot of interests due to energy self-efficiency and sustainable waste management [5]. Septage is usually removed from septic tank by vacuum trucks and transport to a distant treatment plant, however, most of the septage is not well treated and mismanaged leading to environmental problems in the country. Consequently, it requires a suitable method for managing and treatment. Considering a large amount of waste and nutrient-rich characteristic, septage has the potential to be used as substrate to produce biogas. Besides, mixing different wastes with septage has been applied for enhancing biogas production. It is due to the supply of missing and imbalance nutrients by the co-substrate and positive synergisms established in the digestion process that support the growth of microorganisms involved hydrolysis and methanogenesis. Previous research have studied using of septage for anaerobic digestion (AD) as treatment and generated renewable energy as co-digestion with landfill leachate [6], food waste [7], and microalgae [8]. In some cases, septage is used as an alternative fertilizer in agriculture and aquaculture without any prior treatment [9].

However, the performance of the anaerobic digestion process (AD) relies on a combination of physical, chemical and biological processes in which microorganisms play an important role. However, the recent system designs based on the information of microbial community composition still have a limited number and remain unclear due to lacking of sufficient detailed knowledge in understanding of the microbial ecology of the system. There are still much left to be known concerning the underlying mechanisms linking operating conditions such as temperature and substrate of AD systems to microbial community structure and function. Therefore, understanding of the complex microbial communities in the AD and their responses to environmental changes might provide the valuable information that can be used to optimize the AD system [10]. Since 99% of bacterial strains cannot be culture-grown on media, assessing the microbial diversity using molecular techniques have more advantages than the conventional method due to it is rapid, less laborious, more sensitive and specific [11-12]. Previously, many studies have been conducted on microbial communities in the temperature phased anaerobic digestion (TPAD) as denaturing gradient gel electrophoresis (DGGE). Yu and co-workers [13] used denaturing gradient gel electrophoresis (DGGE) for the determination of archaeal community and found *Methanobacteria* and *Methanosarcina* from both mesophilic and thermophilic process. Additionally, Hameed and teams [14] determined the microbial communities in TPAD of municipal wastewater sludge and its bacterial community was dominated by *Firmicutes, Bacteriodetes* and *Proteobacteria* while archaeal community was dominated by *Methanimicrobia* and *Methanobacteria*. It is evidenced that core microbial groups have different growth conditions, physiology and stress tolerance which also varies among waste and system condition. Also imbalance among these organisms due to the disturbances which varied from case to case could cause mulfunction of such system. Hence successful TPAD treating

different types of waste requires the study to elucidate the effect of common operating condition on change of microbial community.

This study aimed to investigate the effect of temperature and substrate on the structure of microbial communities involved in anaerobic co-digestion process between septage and longan peel waste using molecular techniques based on DGGE and metagenomic sequencing. This finding could provide more information of the system and could be applied for further enhancement of the AD system treating agricultural waste.

Methodology

Anaerobic co-digestion process and sludge samplings

Laboratory scale of temperature phase anaerobic digestion systems (TPAD) with two reactors operated at 55°C (thermophilic digester) and 35 °C (mesophilic digester) were conducted by Thunyaluk [15] as shown in **Fig. 1**. The TPAD system was consisted of a 30 L-thermophilic (55˚C) and a 10 L-mesophilic (35˚C) reactors. The Organic Loading Rate (OLR) was started from 0.5/1.0 kg VS/m^3-d (OLR of thermophilic/ mesophilic reactor) and sequentially increased to 4.6/5.0 kg VS/m^3-d, respectively. Longan peeling waste was obtained from dried longan production process in Lumphun province and it was used as a co-substrate mixing with septage. Microbial sludge were sampling from untreated septage (ST) and outlet points of both digestors (LP55 and LP35 for thermophilc and mesophilic digester respectively) every week. They were then preserved by mixing with 70% w/w ethanol with the ratio of 1:1. and stored at -20˚C for molecular analysis using PCR-DGGE technique and metagenomic sequencing.

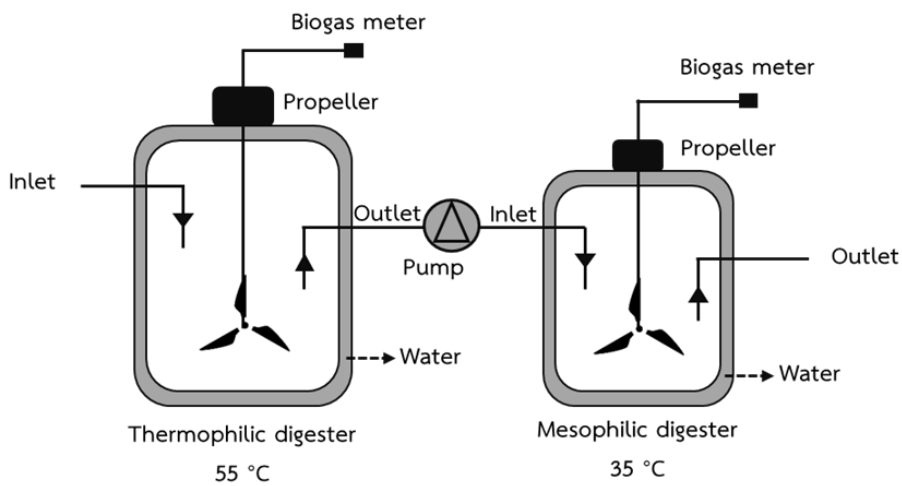

Figure 1 Diagrammatic of temperature phase anaerobic digestion reactor

Microbial community analysis

Total DNA was extracted from the sludge sample using the nucleospin® soil (Bio-rad laboratories, USA) following the manufacturer's instructions. The DNA samples were then sent for metagenomic sequencing analysis which was performed on Illumina HiSeq platform by Novogene Biological Information Technology Co. (Tianjin, China). The relationship of microbial diversity is applied in analyzing the complexity of species diversity for a sample calculated with QIIME (Version 1.7.0), displayed with R software (Version 2.15.3) and generate the Venn and Flower diagram. Unweighted Pair-group Method with Arithmetic Means (UPGMA). Changes of archaea and bacterial communities were analyzed by PCR-DGGE as described by Manatsawat [16] and Pholchan et al [17]. The variable V3 region of 16S rDNA was amplified by using primers targeted to conserved regions of the 16S rDNA gene using specific primers set of 341F/Uni518R [18] and 344F/Arc522R [19]. The amplification conditions used in this study were modified from Hogg and Lehane [20] and conducted using T100 thermal cycler (Bio-rad laboratories, USA). Once the PCR product were stored at -20 °C. DGGE analysis of PCR product was performed on DCode™ system (Bio-Rad laboratories, USA). DGGE images were analyzed via Gene tools analysis software version 3.02.00 of SynGene Genius system (SynGene, UK). Dominant DNA bands representing different bacteria and archaea sequences were excised and purified by RBC TA Cloning Vector Kit (RBC BIOSCIENCE, Taiwan) prior to sequencing (First base, Malaysia) for future study.

Results and Discussions

Performance of TPAD for waste treatment and biogas production

The system performance of anaerobic co-digestion between septage with longan peeling waste are shown in **Table 1**. The results showed that higher biogas production was found from thermophilic digester, while no significant differences ($P \geq 0.05$) of methane contents found from both digesters. Methane production were in the range between 40-56% and 40-58% for thermophilic and mesophilic, respectively. Interestingly, it was found that methane production throughout the operating period from both digesters co-digesting with septage and longan peel waste was more stable than the one feeding with sole substrate (only septage which is explained elsewhere). This possibly could be the result of more complex substances and varieties of substrate composition contained in longan peel waste as co-substrate with septage. Lower COD concentration was found from septage with approximately 11,601.28±9,128 mg/l, while longan peel and septage had higher COD concentration with approximately 38,802±23995 mg/l. Also, high amount of total solid was found from longan peel with the average value of 586,290±24,253 mg/l and C/N of longan peel was about 54%. This is in agreement with some studies suggested that the compositions of the substrates are important for achieving stable processes [21], while low availability of the substrates for microorganisms can be another factor for biomethane production reduction [22].

Table 1 Summary of TPAD reactor performance of anaerobic co-digestion of septage and longan peeling waste

Parameters	Thermophilic digester	Mesophilic digester
pH	7.43 ± 0.28	7.31 ± 0.21
Temperature (°C)	53.62 ± 1.25	35.83 ± 0.87
VFA (mgCH_3COOH/l)	2,020.44 ± 4.88	1,959.10 ± 3.62
Influent COD (mg/l)	38802±23995	46670±8339
Effluent COD (mg/l)	46670±8339	51791±8872
COD removal (% of feed)	27.63±19.02%	18.92±12.92%
Influent TS (mg/l)	85,788±9,637	56,214±10,921
Effluent TS (mg/l)	56,214±10,921	59,273±8,195
TS removal (% of feed)	70.61 ± 1.47	62.86 ± 1.30
Influent VS (mg/l)	76,016±10,477	46,036±8,851
Effluent VS (mg/l)	46,036±8,851	47,791±6,314
VS removal (% of feed)	76.51 ± 1.30	66.35 ± 1.07
Biogas production (ml/day)	10,803.96 ± 41.17	1,730.18 ± 13.99
Total biogas production (ml)	1,761,046.21	282,019.76

Effect of temperature and substrate on relative microbial abundance

The relative abundances of bacterial 16S rRNA transcripts from metagenomic sequencing analysis revealed some differences from the community composition at the DNA level (**Fig. 2**). Firmicutes Bacteroidetes, Cloacimonetes, Tenericutes and Proteobacteria were identified as top dominant phyla within the bacterial community from both digesters and also untreated septage, while their relative abundances varied among samples. This indicated the effect of substrate characteristics and operating condition on microbial community structure. Firmicutes, Bacteroidetes and Proteobacteria were dominated in untreated septage which also appeared in the different abundance ratio in the thermophilic and mesophilic reactors. However, there were some new dominance species harbored in this system. It was found that anaerobic co-digestion decreased the level of Bacteroidetes while increased the level of Firmicutes and relative abundance of Tenericute and Cloacimonetes. This could involve the availability of substances, the metabolic pathway in the biodegradation process and also the operating condition. The results also revealed that high percentage of Firmicutes and Tenericutes were detected from mesophilic reactor, while high ratio of Firmicutes, Bacteroidetes and Cloacimonetes were found from thermophilic reactor. Firmicutes

Bacteroidetes and Proteobacteria have been identified as the main phyla in various AD [23]. Most of members in Firmicutes phylum are syntrophic bacteria that can degrade various VFAs. This coincidence with high percentage of VFA removal in all digesters obtained from this study (50-80%). In addition, Proteobacteria are also important microbes in anaerobic digestion process as they are well-known for glucose, propionate, butyrate, and acetate-utilizing microorganisms [24]. However, all detected bacteria was also able to conduct acidogenesis, which is the second step in the decomposition of organic matter. In addition, Cloacimonetes was decreased only in the mesophilic digester of STLP. However, the relative compositions of these bacteria in digester were variable among different stages of operation which may be associated with the wastewater constituents. According to [25], however, the abundance of phylum Cloacimonetes was linked to lower methane production in the reactors fed with protein-rich substrates. This could explain lower methane production from the digester.

It was found that methanogen communities were unique and diverse between digesters operated with different temperature and substrate variation due to the metabolic pathway occuring in each digesters (**Fig. 3**). The result showed that the euryarchaeota phylum was the most dominant orders. *Methanothermobacter, Methanosarcina, Methanosaeta* and *Methanobacterium* were the top four predominant methanogen genera found from both mesophilic and thermophilic digesters. Some of the methanogens occurring in the system were significantly correlated with methane productions. Some studies suggested that *Methanobacterium* and *Methanothermobacter* was able to grow under high and medium range of temperture, while these species use H_2/CO_2 and formate for methane production [26-27]. Interestingly, *Woesearchaeota* was only found from the thermophilic digester. This species, known as haloarchaea, is distantly related to nitrogen-phosphate remover [28]. This probably linked to large amount of nitrogen-phosphate contained in longan peel in the thermophilic system and may impact on the biological process. In addition, *Methanosaeta* was found to have more population than *Methanosarcina* in septage, whereas *Methanosaeta* was detected less than *Methanosarcina* in TPAD systems co-digested between septage and longan peel. This is because *Methanosarcina* generally have higher growth rates and tolerances against high concentrations of VFA than *Methanosaeta* [29] . From the result, it is likely that some organisms might have only one specific function while some can perform multiple functions such as both hydrolysis and fermentation.

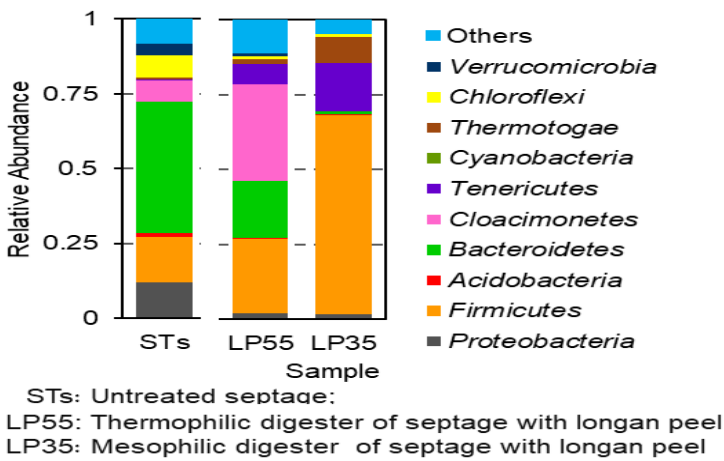

Figure 2 Relative abundance of difference bacteria phylum from TPAD systems at steady state

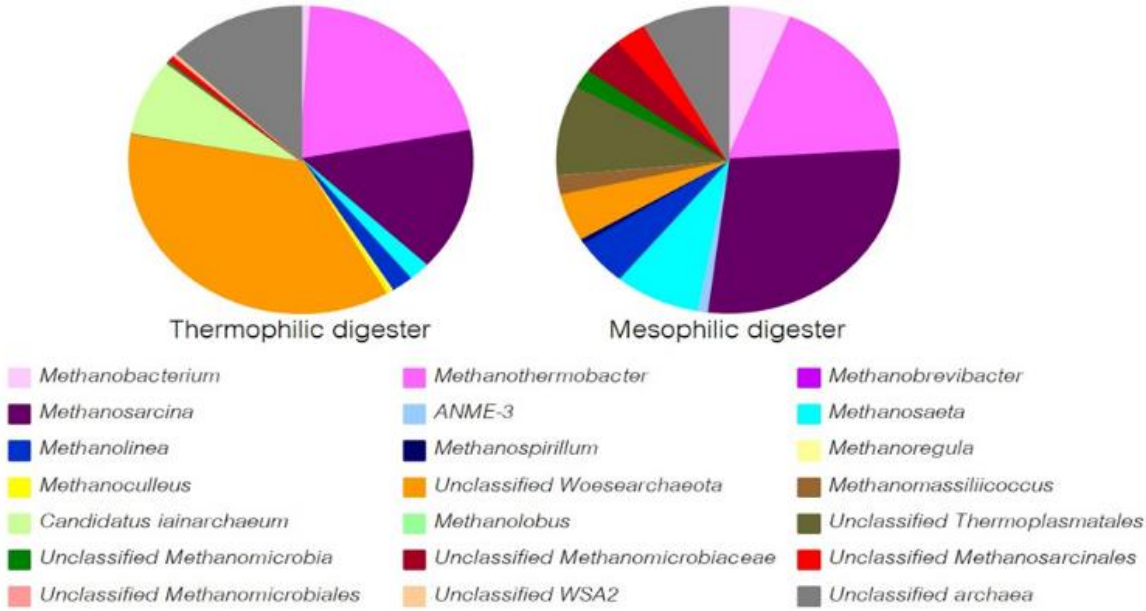

Figure 3 Krona diagram of archaea genera in TPAD system

Effect of temperture and substrate on microbial communities structure

Clustering analysis of bacterial communities from DGGE profile and % relative abundance of dominance species (**Fig. 4 and 5**) showed that thermophilic and mesophilic digester harbored different bacterial communities. There appeared the shifts in the microbial community structure during the operational period. The whole bacterial community and methanogenic community in both digesters formed different groups corresponding to different phase of operation. It was also found that bacterial and archaea community structures were diverse and distinct between digesters due to the variation of substrate and its intermediates and temperature. This coincides with some works reported that substrate variation and temperature had the effect on microbial community structures [30]. Archaea community seemed to be less diverse than bacterial community and the community structure of microbial community in thermophilic reactor showed more stable (**Fig. 5**). This was supported by the Shannon index (*H'*) values which showed that bacterial diversity was higher and have higher similarity than archaea community for both digesters (2.278±0.0605 and 2.268±0.1775 for thermophilic and mesophilic of bacterial community and 1.402±0.092 and 1.589±0.1415 for thermophilic and mesophilic of archaea community). This can be the result of high variety of substrates and intermediate products in the mesophilic phase and there are many different species have the growth condition under this temperature.

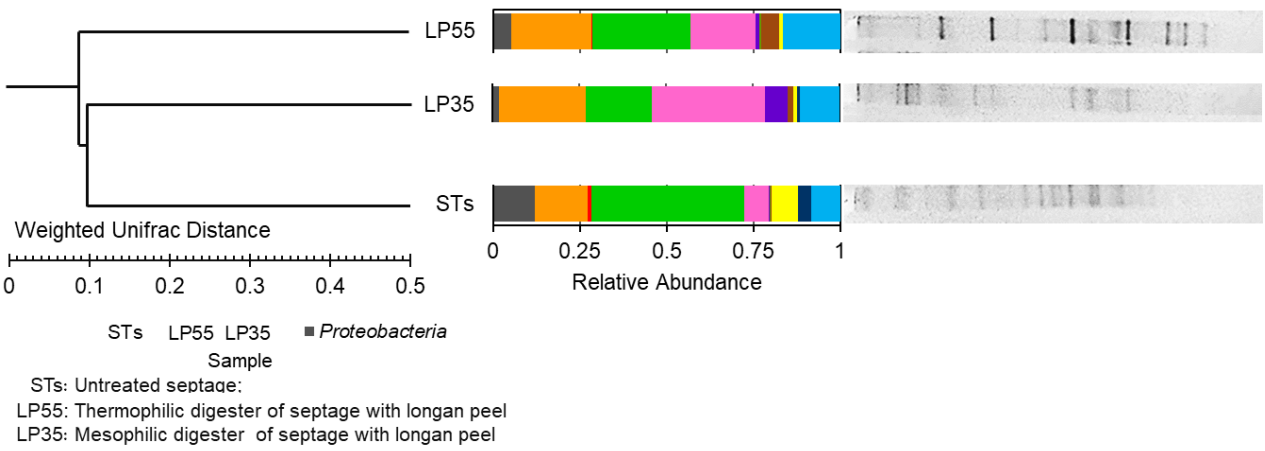

Figure 4 Bacterial DGGE banding pattens from TPAD systems

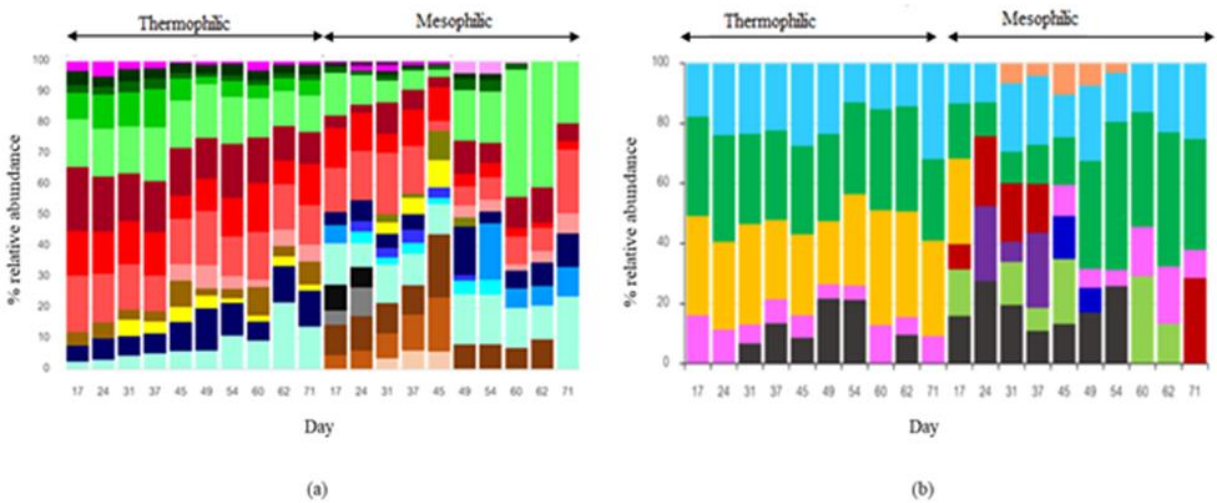

Figure 5 The relative abundance of microbial communities from TPAD system obtained from DGGE profiles (a) bacterial communities (b) Archaea communities

Conclusion

Longan peeling waste and septage were used as the substrate for biogas production in temperature phase anaerobic digestion systems (TPAD) operated under 55°C (thermophilic digester) and 35°C (mesophilic digester). Higher biogas production was achieved from the Thermophilic tank. Each digester harbored distinctive microbial populations, some of which were significantly correlated with the TPAD system performance. The results indicated that *Methanothermobacter* and *Methanosarcina* were the most important methanogenic bacteria, while *Firmicutes Bacteroidetes, Cloacimonetes, Tenericutes and Proteobacteria* were identified as top dominant bacterial phyla from both digesters and also untreated septage. High number of *Firmicutes* and *Tenericutes* were detected from mesophilic tank, while

Bacteroidetes and *Cloacimonetes* were found from thermophilic reactor. Moreover, high ratio of Methanosaeta was found from the system feeding with only septage, whereas it was less detected in TPAD systems co-digested between septage and longan peel. This study proved that substrate and temperture drive the dynamics of key microbial population and its correlation with hydrolytic and metanogenic functionality in the systems.

Acknowledgement

This work was partially supported by Thailand national research funding number MJU1-61-039, Maejo University's disciple scholarship and STEM Workforce from NSTDA.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 23-32
www.eeat.or.th

Indoor Air Quality of PM2.5 in Classrooms of Science Building, Udon Thani Rajabhat University, Thailand

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Abstract

This work investigates for indoor air quality particulate matter 2.5 microns (PM2.5) in classrooms of Science building at the Faculty of Science, Udon Thani Rajabhat University, Thailand. The study was determined from selected five classrooms and monitored by a dust track meter during the summer (April-May 2019) and rainy season (June–July 2019). PM2.5 concentrations in summer were found between 53.1±11.10 μg/m^3, which over the maximum acceptable value PM2.5 for indoor air quality in office buildings (8 hours) by the Ministry of Public Health, Thailand (35 μg/m^3), while in the rainy found only 25.6±4.99 μg/m^3. This indicated summer was also possibly related to the thermal inversion phenomena, which often occurs in high and low temperatures between indoor and outdoor sites. The results showed the average PM2.5 concentration in laboratory rooms (43.2±9.36 μg/m^3) was higher than that of the lecture rooms (32.71 μg/m^3). This is because the activity of class occurred during the laboratory lesson time. Additionally, the outdoor source caused by air pollution from sugar cane combustion near Udon Thani University can be generated the high PM2.5 (Average 91.70±21.62 μg/m^3). As because of this, PM2.5 generated by particles of these activities was the critical environmental problems in the Udon Thani city. The results show that new data in Udon Thani university for the air quality (PM2.5) based on concentrations from indoor air in classroom, including caused of PM2.5 pollution is emitted from outdoor pollution sources. It is well known that air pollution constitutes the most pressing environmental health risk and environmental problems. Therefore, the data of this study to benefit for the local government and environmental organizations to reduce or prevent the concentrations of many pollutants are influenced by local in Udon Thani city.

Keywords : Indoor Air Quality; PM2.5; Air Pollution Monitoring; Classroom; Udon Thani

Introduction

Particulate matter 2.5 micron (PM2.5) is a major air pollution problem that can be contributed to the toxic and biological pathogen city [1]. PM2.5 can cause harm to the human respiratory system and deposits in alveolar regions [2]. The component of PM2.5 is well known generated by combustion during the waste combustion or using fireplaces [3, 4]. On the other hand, PM2.5 from outdoor sources originated mainly from internal combustion engine exhaust soot [5]. Thailand is the world's 4th largest sugarcane producer and the 2nd largest sugar exporter, while Udon Thani province is the major producer for a big sugar exporter in Thailand [6]. Udon Thani is located at the northeast of Thailand and serves as a centre of transportation and economic, especially the sugar industry [7]. Sugar cane is essential raw material for sugar industries, so it is important for problems effect of the environment [8]. The percentages of burned sugarcane in the production found higher and mainly caused by air pollution in the forms of smoke, toxic gas, dust, and particles in the air [9].

At present, the sugarcane industry's trend to expand every year, sugarcane farmers grow more sugarcane and harvest manually in most areas. For convenience, sugar cane farmers choose to burn sugarcane before harvesting, this burning prior to harvesting causes air pollution [10]. As because of this, PM2.5 generated by particles from the sugar cane burning is one of the most critical environmental problems in the Udon Thani city. The agricultural biomass burning is the major source of outdoor air pollution in Udon Thani province which is seasonal from January to April in every year. The situation could be worse in the future unless proper measures are undertaken. More recently, PM2.5 is well known negative health effects on humans and can lead to long-term and short-term health problems [11]. Specially, students in universities always spend more time in classrooms which can increase their inhaled doses of indoor air pollution [12, 13]. This is mostly air pollution, contaminated in the classrooms of the university such as allergens, particles, volatile organic compounds [14]. Moreover, a high concentration of PM2.5 in indoor air may cause acute or chronic health effects, and even cause premature deaths in the elderly people and people with asthma [15, 16]. Thus, the indoor air quality of PM2.5 should be studies became an important research topic for public health. Furthermore, the World Health Organization (WHO) also reports published the limit on indoor air pollution level of PM2.5 including the particularly in relation of classrooms in schools or universities is still limited [11, 17].

Therefore, the aim of this work was investigated the indoor air quality of PM2.5 in the university classrooms during lessons. The objective was to determine the concentration of PM2.5 including the temperature, and relative humidity was studied in five classrooms, which is located on the 6th floor of the Science building, Faculty of Science, Udon Thani Rajabhat University. Experimental method investigated during in the summer (April-May 2019) and rainy season (June-July 2019). The concentrations of particulate matter were determined by a DustTrak™ aerosol meter with using a light scattering principle. This method is wildly used for indoor air quality monitoring in office buildings following the methods reported in previous works [3, 4]. The level of PM2.5 concentration between different seasons and various floors of Science building was determined and compared. Finally, the obtained monitoring of indoor air quality PM2.5 including the maximum, minimum, and average PM2.5 concentrations in classrooms of the Udon Thani University was promoted to improve this situation or environment management in the future.

Methodology

Area sites

This work was studied in five classrooms by the purposive sampling of Science building (ScB) of the faculty of Science, Udon Thani Rajabhat University (Sam Phrao campus), where is in Udon Thani province. Udon Thani Rajabhat University has two main campuses, and the faculty of Science is in Sam Phrao campus (N 17.450816, E 102.936165). More information, the faculty of Science has four buildings, namely ScB1, ScB2, ScB3 and ScB4. The selected building on this work was selected from ScB1 (6^{th} floors). The indoor air quality of PM2.5 was carried out in the summer (April to May 2019) and rainy season (June to July 2019) to compare. The average data of indoor temperature, and relative humidity was also carried out.

Classroom sampling sites

The classrooms of Science building 1 (ScB1) in this work were divided by lecture room and laboratory room types. To compare the high PM2.5 concentrations during the activity in each classroom, the lecture room was studied for no class activity or movement, while in the laboratory room studied for class activities and movement. Indoor air quality data were collected from the ScB1 building (6^{th} floors). The classroom sampling was selected from each floor by the randomly sampling from 80% of the total floors of the building [18]. Therefore, the five selected classrooms were collected from 1^{st} - 5^{th} floor. For the limitations of monitoring instruments and class activity, this work monitored indoor air for PM2.5 concentrations in the selected five classrooms for once a week of one classroom and per one month. The five of selected classrooms from ScB1 were namely $ScB1_1$, $ScB1_2$, $ScB1_3$, $ScB1_4$ and $ScB1_5$ following the classroom from 1^{st} floor, 2^{nd} floor, 3^{rd} floor, 4^{th} floor and 5^{th} floor, respectively, as shown in **Figure 1**.

The period time in this work was investigated on a working day with 8 hours (08.00 a.m. to 16.00 p.m.) to compare with the maximum acceptable value for indoor air quality in office buildings that PM2.5 must not exceed 35 μg/m^3. All the lecture room type ($ScB1_1$ and $ScB1_2$) had a same capacity of 40 persons and a volume of 192 m^3, while all the laboratory room type ($ScB1_3$, $ScB1_4$ and $ScB1_5$) had a same capacity of 60 persons and a volume of 384 m^3. All classrooms were equipped with the standard school tables, chairs, fans, and a blackboard with chalk at the front. In addition, the selected classrooms of ScB1 building were naturally ventilated by opening windows all time in the lecture with the class activity. All classrooms in this studied had no air conditioning and open all windows during the monitoring time.

Sampling of PM2.5 by a DustTrak meter

PM2.5 was determined by a DustTrak meter with a model 1103 (TSI Incorporated, USA). PM2.5 was monitored and measured by a DustTrak meter with flow rate 3±0.01 L/min based on of a light scattering method. The particle of PM2.5 concentrations was measured with a real-time in each classroom and obtained with a 1-minute time resolution. The instrument was calibrated daily to a zero filter that used to zero setting as a unit and ensure for accuracy of reading. The DustTrak meter was placed in a middle back corner of the classroom about 1.0 m above the floor, which corresponds to the breathing zone of the sitting student, as illustrated in **Figure 2**. To study indoor air quality that influenced by and therefore related to outdoor air quality, the PM2.5 concentration was also measured. The DustTrak meter was placed in a front of ScB1 building about 1.5 m above the floor and 1.0 m from the roadside, which corresponds to the ambient air monitoring.

Figure 1 The selecting classrooms of Science building (ScB1) with each floor

Figure 2 The sampling of PM2.5 by a DustTrak meter in classrooms

Results and Discussion

Temperature and relative humidity in classrooms

A monitoring of indoor air quality PM2.5 from the selected five classrooms in the Science building (ScB1), Faculty of Science, and Udon Thani Rajabhat University was investigated the temperature (T) and the relative humidity (RH) as shown in **Table 1**.

A summary detailed of temperature and relative humidity in the classrooms from April to July 2019 is divided into summer and rainy seasons as shown in Table 1. Thailand is a most of the rains in the June to September that considered as the rainy season, while January to early May is considered as summer of the year. The average of temperature and relative humidity in the selected five classrooms were 30.96±2.63℃, RH=72.0±8.6%, respectively. Obviously, the average temperature in

classrooms in summer found 33.06±2.12°C, while in rainy found 28.85±0.48°C. For the average relative humidity was also the same trend, it was found 64.1±4.4% in summer, while in rainy found 79.8±1.2%. It was observed that the temperature and relative humidity in summer were higher than that of in rainy. As the results, it was considered to discuss for the factors of dust re-suspension in the next discussion.

Table 1 Temperature and relative humidity values of the selected five classrooms in winter and rainy season

Floor of building	Name	Room type	Summer (2019)				Rainy season (2019)			
			April		May		June		July	
			T (^{o}C)	RH (%)	T (^{o}C)	RH (%)	T (^{o}C)	RH (%)	T (^{o}C)	RH (%)
1	$ScB1_1$	Lecture	35.5	61	31.5	68	29.5	81	29.0	80
2	$ScB1_2$	Lecture	34.7	60	30.7	69	29.0	80	28.3	81
3	$ScB1_3$	Laboratory	35.0	59	30.5	69	28.7	82	28.0	78
4	$ScB1_4$	Laboratory	35.0	60	31.0	67	29.0	79	28.5	79
5	$ScB1_5$	Laboratory	35.0	60	31.7	68	29.5	79	29.0	79
Average of month			35.04	60.0	31.08	68.2	29.14	80.2	28.56	79.4
Average of season			T = 33.06±2.12°C, RH = 64.1±4.4%				T = 28.85±0.48°C RH = 79.8±1.2%			
Average of classrooms			30.96±2.63°C, RH=72.0±8.6%							

PM2.5 levels in classrooms

The average PM2.5 concentrations obtained from a real-time monitoring of 8 h from the selected five classrooms. In the summer, it was found that the average PM2.5 concentration were 53.1±11.10 μg/m^3, while in the rainy season only 25.6±4.99 μg/m^3. For comparison of month, the results showed the average of PM2.5 concentrations in ranged from 63.0, 40.8, 26.6 and 24.6 μg/m^3 in April, May, June, and July (2019), respectively. When comparing the different type room with the same students (40 persons), it was differences for observed for the average of PM2.5. The PM2.5 concentration in a laboratory room type (43.2±9.36 μg/m^3) was higher compared with a lecture room type (32.71 μg/m^3). This result indicated that the concentration of PM2.5 was increased in summer season, especially in April 2019 (63.0±10.60 μg/m^3) that observed in the highest temperature (35.04°C). The higher PM2.5 concentrations in summer were also possibly related to the thermal inversion phenomena, which frequently occurred in high and low temperature. The maximum acceptable value for the indoor air PM2.5 in office buildings is limited in 8h at 35 μg/m^3 [18]. As the results, the average PM2.5 concentration in this work (April to July 2019, monitored 8h per day) was found 38.75±12.81 μg/m^3 that over limited of the maximum acceptable value for the indoor air PM2.5 in office buildings. A high PM2.5 concentration was found during class activities from the laboratory room type and ground floor of building that should be focus. Therefore, it should be beware and continue monitored to find environmental management in this situation.

Generally, a high PM2.5 level was found in the classrooms indicating the particles related to major PM2.5 indoor sources such as blackboard dust and organic matter of students as well as the re-suspension phenomena of particles. However, many previous reports [12, 17] .were found the outdoor sources also main factor to increase the level of PM2.5 in the classroom, such as the vehicle's combustion, the burning industries or the product burning from agriculture. Udon Thani province is the major producer for a big sugar cane exporter and sugar cane burning process was mainly caused to the air, hence, it is important for problems effect of the environment. Especially for sugarcane farmers in Sam Phrao in Udon Thani province, their mostly burned the sugarcane in December to April in every year and it obtained to the toxic gas, dust, and particles that caused to problem of air pollution. The biomass activities from the burning sites related to the university was found in the working time of day (07.00 a.m.-17.00 p.m.). The approximately size of burning was 2500 rai. As because of this, PM2.5 generated by particles from the sugar cane burning is one of the most critical environmental problems in the classroom of Udon Thani Rajabhat University.

According to Table 2, the average of PM2.5 in summer generated from outdoor source was found at 129.2±17.26 μg/m^3, while in rainy was found at 54.2±10.23 μg/m^3. Obviously, these results could be explained that in the summer (April to May 2019) showed the higher PM2.5 more than in the rain (April to May 2019) as shown in Figure 3. Moreover, April 2019 was observed the highest PM2.5 concentration (63.0μg/m^3) that caused from outdoor sources (159.8μg/m^3) by sugar cane burning and the high temperature (35.04°C). In the rainy season, it has the rain effect that can be reduced the particle in the ambient air from the outdoor source. This is the effect of the low PM2.5 concentration in June and July 2019. Additionally, the higher levels of particle concentration found in summer were in accordance with the corresponding to previous studies [14, 15].

Table 2 Summary of the average PM2.5 concentrations in selected five classrooms

Floor of building	Classroom	Description	Room area (m^2)	PM2.5 concentrations (μg/m^3)							
				Summer (2019)				Rainy (2019)			
				April		May		June		July	
				Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
1	$ScB1_1$	Lecture	192	55	126	37	88	24	50	22	44
2	$ScB1_2$	Lecture	192	48	110	33	70	20	46	18	43
3	$ScB1_3$	Laboratory	384	82	227	59	127	34	81	32	70
4	$ScB1_4$	Laboratory	384	62	155	39	117	27	53	26	48
5	$ScB1_5$	Laboratory	384	68	181	36	91	28	58	25	49
Average of month				63.0	159.8	40.8	98.6	26.6	57.6	24.6	50.8
Average of season				Indoor air 53.1±11.10 μg/m^3				Indoor air 25.6±4.99 μg/m^3			
				Outdoor air 129.2±17.26 μg/m^3				Outdoor air 54.2±10.23 μg/m^3			
Average of indoor air pollution				38.75±12.81 μg/m^3							
Average of outdoor air pollution				91.70±21.62 μg/m^3							

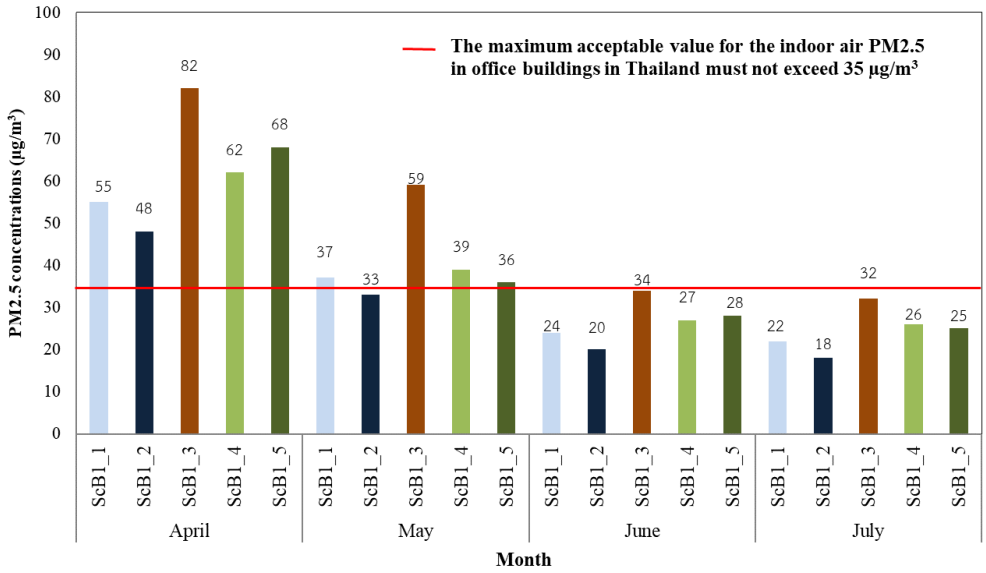

Figure 3 The concentration of PM2.5 from five classrooms for each month

The indoor air quality of PM2.5 concentration measured from five classrooms was focused within each class of the month. Considering for each month, the result indicated the PM2.5 concentrations from a laboratory room ($ScB1_3$, $ScB1_4$ and $ScB1_5$) was higher more than a lecture room ($ScB1_1$ and $ScB1_2$). The high PM2.5 concentrations were found during the activity class in each day, while in the lecture room no class activity or movement. Thus, the PM2.5 concentrations from a lecture room were observed less. This is because of a major classroom activity occurred, rather than during class time when the student was seated at their tables. Moreover, a high concentration of PM2.5 was also detected in the higher floor more than that of the first floor. This is because during break time, students were movement for going in and out of the classrooms. Furthermore, it was obvious that the average PM2.5 concentration was higher in the front of classrooms than in the rear of the classrooms that caused by a fine particle from backboard. These indicated that one possible reason for PM2.5 pollution in the classroom was the re-suspension of settled particles due to the class activity of the student movement. More understandingly, this study found the indoor particle number concentrations at classrooms were related to the high activity times, and higher in summer than in rainy, according to the previous studies [4, 12]. The maximum PM2.5 concentration was observed after studying after 1 h in the morning and afternoon, while the minimum PM2.5 also observed in lunchtime and after studied over 3 h, as illustrated in **Figure 4**. It is difficult to make a direct comparison, because of different sampling time and measuring instrument used between studies.

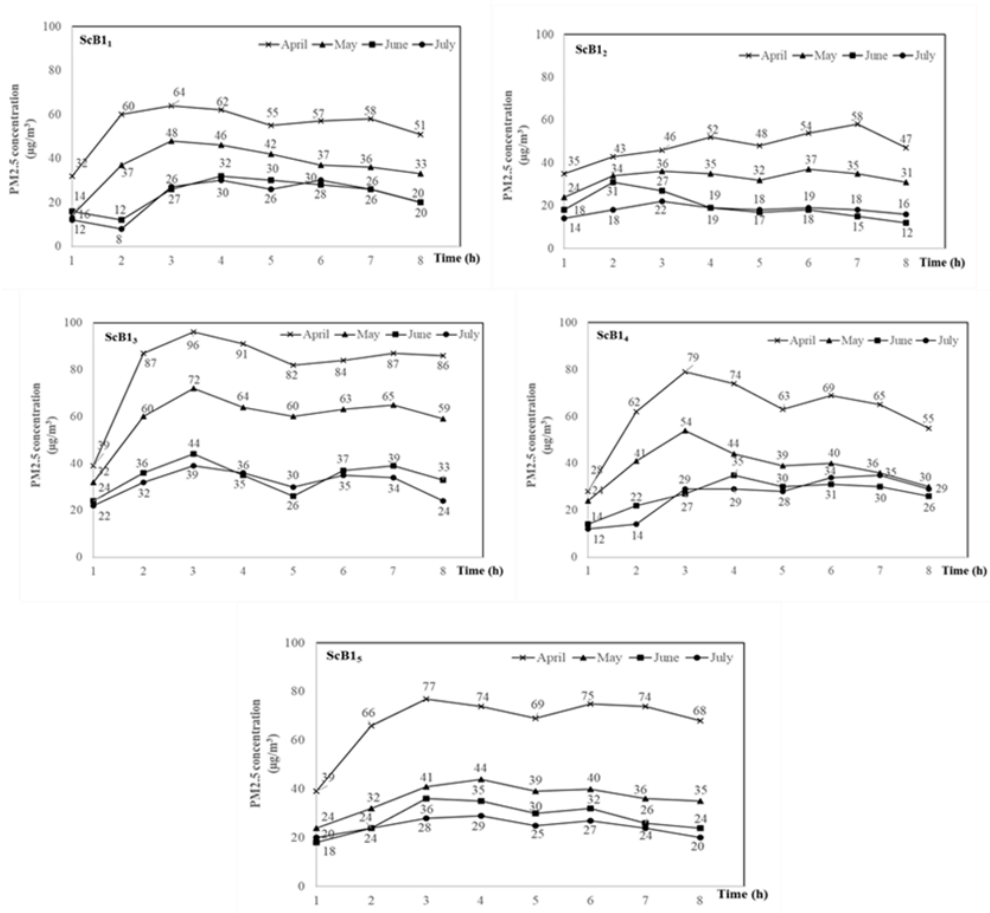

Figure 4 A real-time 8h for PM2.5 concentration monitored from each classroom

Conclusion

The study was determined from five classrooms within each level of the Science building. In summer found the higher PM2.5 concentration more than that of in rainy. This is because the activities from class, sugar cane burning pollution from the outside and meteorological in high temperature. During the rain, the windows were closed during class studied to prevent rain and dust from entering classrooms. In the summer, the windows were open most of the time, in this case the high indoor particular matter levels observed in our study could be explained by infiltration from the outdoor environment and the movement of student studied in classrooms. It can be indicated that distribution of PM2.5 concentration was increased in summer season. Moreover, a high concentration of PM2.5 from difference five classrooms was dependent on class activity occurred, rather than during class time when the student was seated at their tables. A laboratory room found PM2.5 concentration higher that of a lecture room. This is because during break time, students were movement for going in and out of the classrooms, and recreational activities inside the building with different floor. The results in this study show that air quality of PM2.5 based on

concentrations of the indoor air in classrooms at Faculty of Science, Udon Thani Rajbhat University, including how much of each pollutant is emitted from various pollution sources. It could be observed that outdoor air pollution is the one of the health and environmental problems. The results showed the concentrations of PM2.5 pollutants are influenced by local. Therefore, the indoor air quality in this work is influenced by and therefore related to outdoor air quality. In addition, the data of this study the new data of air pollution in Udon Thani province including outdoor air pollution impact to health student that could be benefit for the local government and environmental organizations to enforce to reduce or prevent the concentrations of many pollutants are influenced by local in Udon Thani city.

Acknowledgement

The authors are grateful to thank Asst. Prof. Adisak Bootwong and Dr. Winai Meesang for their equipment support. The authors wish to thank Mechanical Engineering, Faculty of Technology, Udon Thani Rajabhat University and Faculty of Sciences and Technology, Suan Sunandha Rajabhat University for validation of analytical methods.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 33-44
www.eeat.or.th

Heavy Metals in Sediments and Water at the Chao Phraya River Mouth, Thailand

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Abstract

Sediment and water samples collected from 20 sampling stations located in the Chao Phraya River Mouth, Thailand during 2017 and 2019 were analyzed for concentrations of mercury (Hg) as well as other heavy metals including cadmium (Cd), zinc (Zn), nickel (Ni) and lead (Pb). The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in sediments of the Chao Phraya River Mouth during 2017 and 2019. The higher concentrations of heavy metals were found in the suspended sediment in comparison to the bottom sediment. The concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in water of the Chao Phraya River Mouth during 2017 and 2019 were lower than the coastal water quality standard in Thailand. Because of the high freshwater inflow and the discharge of low amount of heavy metals in the treated industrial effluent to the Chao Phraya River, the high levels of heavy metals in sediments of the Chao Phraya River Mouth might be caused by weathering of rock, sediment transport and deposition within the Chao Phraya River Basin. The spatial variation of heavy metals in sediments of the Chao Phraya River was existed, due to adsorption and co-precipitation of dissolved heavy metals onto sediments and aggregation-sedimentation of suspended sediment during transport. The seasonal variation of dissolved heavy metals in water of the Chao Phraya River Mouth was observed, due to accumulation and release of dissolved heavy metals from sediments.

Keywords : Heavy metals; Source identification; Water; Sediment; Suspended sediment; the Chao Phraya River Mouth

Introduction

In Thailand, the Chao Phraya River, which is the largest and most important river, passes through 10 cities, including Nakhon Sawan, Uthai Thani, Chai Nat, Singburi, Aug Thong, Ayutthaya, Pathum Thani, Nonthaburi, Bangkok, and Samut Prakan. In the Chao Phraya River Basin, many developments with high rate of urbanization and industrialization along the river banks and south of the Bangkok Metropolis, as well as agricultural modernization along the northern section of the river have been proceeding continuously over the last decade. Such developments possibly discharge wastewater or non-standard effluent to the river, thereby deteriorating aquatic environment and natural resources [1, 2]. Wastes of agrochemicals, urban areas and numerous industries, such as electroplating and electronic equipment, located along the river and large canals connecting the river, either partially treated or without treatment, are discharged into the Chao Phraya River, the Chao Phraya River Mouth and eventually into the Upper Gulf of Thailand causing water quality deterioration with pollutants, including heavy metals [3].

In the Chao Phraya River Mouth, it has been faced the problem from heavy metals. A survey by Polprasert, 1982 [1] found that the accumulation of Cd and Pb in water of the Chao Phraya River Mouth was significant. Wijaya et al., 2013 [4] indicated that the high concentrations of heavy metals (Cd, Cu, Cr, Pb and Zn) in sediment of the Chao Phraya River were observed, reflecting primarily lithogenic baseline concentration of heavy metals in unpolluted sediment. Qiao et al., 2015 [5] found that the concentrations of heavy metals (Cu, Pb, Zn, Cr and Cd) in sediment of the Chao Phraya River were higher than that of the Bang Pakong River, the Tha Chin River and the Mae Klong River. Mclaren et al., 2004 [3] reported that temporal variation of cadmium concentration in water and sediment of the Chao Phraya River was observed, most probably related to seasonal high water levels and sediment flushing.

In the aquatic environment, contamination of heavy metals is a serious threat due to its abundance, accumulation, persistence and environmental toxicity. Both natural actions and anthropogenic activities are responsible for the abundant of heavy metals in the aquatic environment. The heavy metals can be accumulated and magnified to mussels, oysters, shrimps and fish, and can be transferred to humans with the food chain pathways. The increasing pollutions by heavy metals have a significant adverse health effects for invertebrates, fish and humans [6-8].

The principal comportment of metals is a function of the water chemistry and sediment in the natural water body. During transportation of heavy metals in the riverine system, it might undergo frequently changes due to dissolution, precipitation and sorption phenomena [6]. The heavy metal concentrations in sediment are extremely higher than that in water because heavy metals tend to accumulate in bottom deposits [7]. However, heavy metals amassed in bottom sediment are more likely to be re-suspended and cause secondary contamination of heavy metals to the water column [8].

The aims of this study were (i) to monitor heavy metals (Hg, Cd, Pb, Ni and Zn) in sediments and water of the Chao Phraya River Mouth, Thailand during 2017 and 2019, (ii) to assess contamination levels of heavy metals, seasonal and spatial variation of heavy metals in sediments and water of the Chao Phraya River Mouth and (iii) to identify possible sources of

heavy metals in sediments and water of the Chao Phraya River Mouth.

Methodology

Study area and sampling

Sediment and water samples were collected from 20 sample locations of the Chao Phraya River Mouth (**Figure 1**). Sampling was performed in four phases: firstly, January 2017 (dry season); secondly, June 2017 (wet season); thirdly, July 2018 (wet season) and fourthly, January 2019 (dry season).

A total of 160 samples (80 sediment samples and 80 water samples) were collected. About 100 g of surface sediment samples (15 cm depth) and 1 L of surface water samples were collected from 20 sampling stations of the Chao Phraya River Mouth, using a sediment collector and a grab method, respectively during the lowest tide. After collection, sediment and water samples were acidified with nitric acid (pH<2), stored at 4°C in the dark and transferred to the laboratory.

Sample digestion and analysis of samples

The sediment, and filtered and non-filtered water samples were digested with HNO_3-HF-HCl acid. 1 g of each sample was weighted into microwave vessels and digested with 3 mL 65% HNO_3, 1 mL 40% HF and 0.8 mL 37% HCL. The digestion was carried out using a microwave digester for 26 min. The digests were cooled to room temperature, filtered through a 0.2 µm membrane filter and diluted to 20 mL with distilled water. The filtrate from sediment samples was also analyzed for total heavy metals (Hg, Cd, Pb, Ni and Zn) using ICP-OES. The filtrate from filtered and non-filtered water samples was analyzed for dissolved and total heavy metals (Hg, Cd, Pb, Ni and Zn), respectively using ICP-OES. Difference of total and dissolved heavy metals in water was reported as values of heavy metals in suspended sediment.

Results and Discussion

Heavy metal concentration in bottom sediment

The mean values of heavy metals in sediment monitored at the Chao Phraya River Mouth, Thailand on four times during 2017 and 2019 (January 2017; June 2017; July 2018; January 2019) are summarized in **Figure 2**. The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in sediment of the Chao Phraya River Mouth during 2017 and 2019. The concentrations of lead (Pb), zinc (Zn) and nickel (Ni) in sediment monitored in January 2017 were similar to those concentrations in sediment

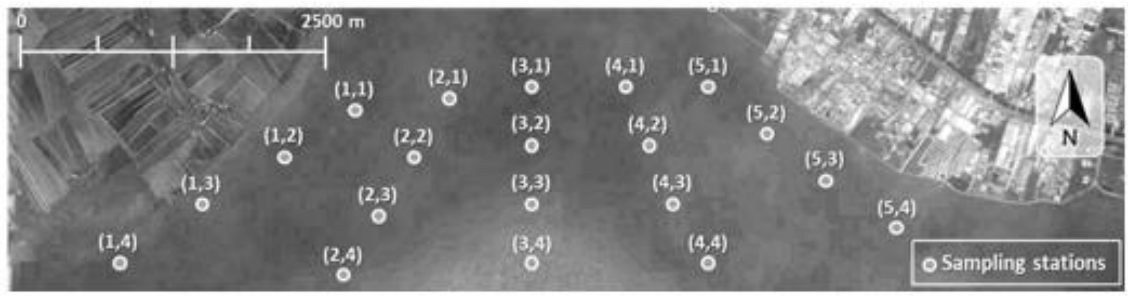

Figure 1 Sampling stations of sediment and water at the Chao Phraya River Mouth

monitored in July 2018 and January 2019. The mean concentrations of lead (Pb), zinc (Zn) and nickel (Ni) in sediment monitored in January 2017, July 2018 and January 2019 were 0.010-0.014 mg/kg for Pb, 0.017-0.022 mg/kg for Zn and 0.002-0.006 mg/kg for Ni. The lowest concentrations of cadmium, Cd (0.0003-0.0005 mg/kg) were accumulated in sediment monitored during 2017 and 2019. The mean concentrations of heavy metals in sediment monitored during 2017 and 2019 decreased on the following order: Zn > Pb > Ni > Cd. On the other hand, the mean concentrations of mercury (Hg) in sediment decreased significantly with increasing periods of monitoring. According to Menasveta and Cheevaparanapiwat, 1981 [9] and Hungspreugs and Yuangthong, 1983 [10], the high concentrations of lead (Pb) and mercury (Hg) were found in bottom sediment and green mussel of the Chao Phraya River Mouth.

Figure 2 indicates that the concentrations of mercury (Hg), zinc (Zn), and lead (Pb) in sediment of the Chao Phraya River Mouth in June 2017 and July 2018 in wet season were lower than those concentrations in January 2017 and January 2019 in dry season. According to Mclaren et al., 2004 [3], cadmium concentrations in sediment of the Chao Phraya River in wet season were lower than those concentrations in dry season. This might be due to flushing of sediment by large volumes of rapid moving water during wet season. On the other hand, Sirirattanachai and Utoomprurkporn, 2005 [11] found that mercury concentrations in sediment collected from 94 km upstream to the Chao Phraya River Mouth in wet season were higher than those concentrations in dry season, possibly due to re-suspension, suspended sediment transport and deposition.

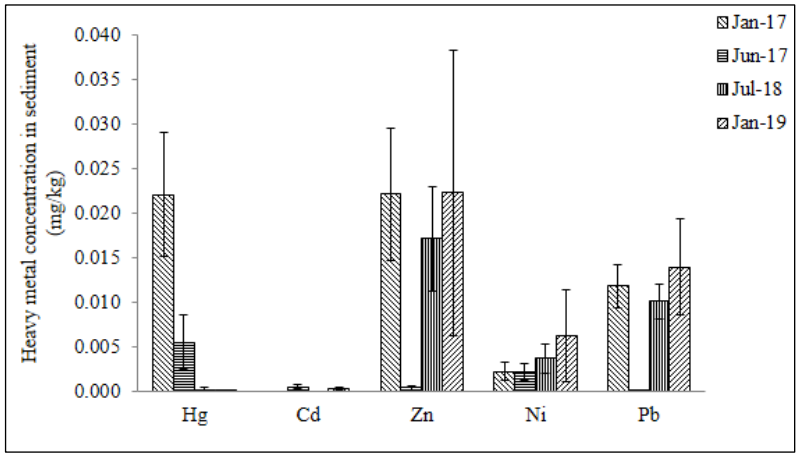

Figure 2 Mean values of heavy metals in sediment in the Chao Phraya River Mouth during 2017 and 2019. Each error bar indicates the standard deviation of sediment samples collected from 20 sample locations of the Chao Phraya River Mouth.

The concentrations of heavy metals in sediment of the Chao Phraya River Mouth, Thailand with other World rivers and river mouths are presented in **Table 1**. The results showed that the values of heavy metals (Hg, Cd, Zn, Ni and Pb) in sediment monitored at the Chao Phraya River Mouth, Thailand during 2017 and 2019 were detected in very low concentrations compared to sediment of other World rivers and river mouths. The seasonal and spatial variations of heavy metals in sediment of the Chao Phraya River, Thailand, the Liuyang River, China, the Karnaphuli River, Bangladesh and the Ghaghara River, India and in sediment of the Chao Phraya River Mouth, Thailand and the Yangtze River Mouth, China were significant [4, 6, 12-14]. Wijaya et al., 2013 [4] reported that river sediments collected from 72 km upstream to 27 km downstream from the Chao Phraya River Mouth showed spatial variation of heavy metals (Cd, Cu, Cr, Pb and Zn), possibly due to adsorption and co-precipitation of dissolved heavy metals onto suspended and bottom sediments, and aggregation-sedimentation of suspended sediment during transport.

The suspended sediment in the river might be caused by weathering of rock through water and wind which is continuously transported by river flow, tide and seasonal monsoon, and deposited along the river flow slow down, especially at the inside of river bends, and river mouths [5]. Because of the high freshwater inflow in the Chao Phraya River and the discharge of low amount of heavy metals contained in the treated industrial effluent into the Chao Phraya River, possible sources of heavy metals in sediment of the Chao Phraya River were not anthropogenic, but rather lithogenic. Wijaya et al., 2013 reported [4] that the values of heavy metals detected in sediment of the Chao Phraya River represented mainly lithogenic baseline concentration of heavy metals in the unpolluted sediment.

Heavy metal concentration in suspended sediment

The mean values of heavy metals in suspended sediment monitored at the Chao Phraya River Mouth in July 2018 and January 2019 are shown in **Figure 3**. The results indicated that the enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in suspended sediment of the Chao Phraya River Mouth during periods of monitoring. The mean concentrations of heavy metals in suspended sediment of the Chao Phraya River Mouth in July 2018 and January 2019 decreased on the following order: Pb > Ni > Zn > Cd > Hg. The high contamination levels of heavy metals in the river mouth suspended sediment might be caused by weathering of rock through water and wind which are natural sources of heavy metals in the environment, and the suspended sediment transport to the river mouth by river flow, tide and seasonal monsoon [15, 16]. Besides natural origin, the variation of heavy metals in the river mouth suspended sediment may be due to adsorption and co-precipitation of dissolved heavy metals onto the suspended sediment.

Table 1 Heavy metals in sediment of the Chao Phraya River Mouth, Thailand and other World rivers and river mouths

Metals	Chao Phraya River Mouth Sediment, Thailand	Chao Phraya River Sediment, Thailand		Liuyang River Sediment, China	Yangtze River Mouth Sediment, China	Karnaphuli River Sediment, Bangladesh	Ghaghara River Sediment, India
		72 km	27 km				
Hg, mg/kg	0.0001-0.022				0.065		
Cd, mg/kg	0.0003-0.0005	0.13	0.22	1.24	0.130	0.63-3.56	0.21-0.28
Zn, mg/kg	0.0004-0.022	21.9	103	138.48	71.5		13.26-17.59
Ni, mg/kg	0.002-0.006			17.48			15.29-25.59
Pb, mg/kg	0.0001-0.014	15.1	28.8	37.82	25.8	21.98-73.42	10.71-14.26
Reference	This study	[4]		[13]	[12]	[6]	[14]

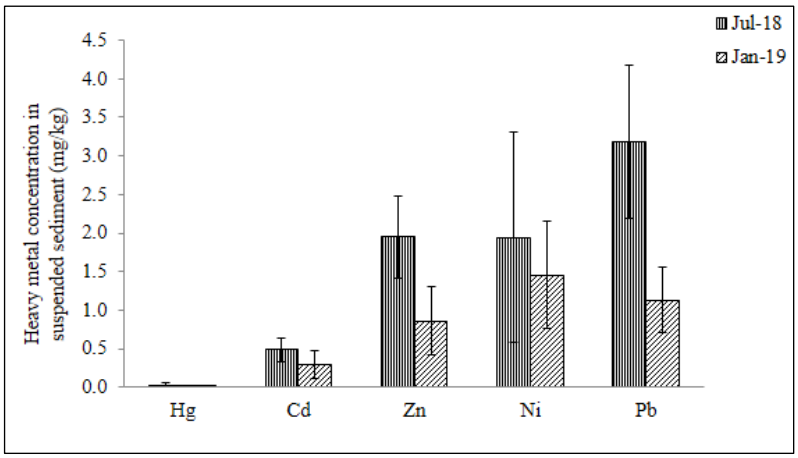

Figure 3 Mean values of heavy metals in suspended sediment in the Chao Phraya River Mouth during 2018 and 2019. Each error bar indicates the standard deviation of suspended sediment samples collected from 20 sample locations of the Chao Phraya River Mouth.

Figure 3 shows that the mean concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in suspended sediment of the Chao Phraya River Mouth in July 2018 in wet season were higher than those concentrations in January 2019 in dry season. The seasonal variation of heavy metals in the river mouth suspended sediment might be due to the fast river flow in wet season, result in more suspended sediment transported to the river mouth. Furthermore, Figure 2 and Figure 3 indicate that the mean concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) in the river mouth suspended sediment were higher than those concentrations in the river mouth bottom sediment. The results of this study were similar to the results of Sabri et al., 1993 [17] and Asokbunyarat and Sirivithayapakorn, 2017 [18]. The low contamination levels of heavy metals in the river mouth bottom sediment might be due to dilution with organic matter and other pollutants.

Heavy metal concentration in water

The mean values of dissolved heavy metals in water monitored at the Chao Phraya River Mouth on four times during 2017 and 2019 (January 2017; June 2017; July 2018; January 2019) are presented in Figure 4. The mean values of dissolved heavy metals in water monitored in January 2017, which decreased on the following order: Pb > Zn > Ni > Cd > Hg, were higher than that of heavy metals monitored in June 2017, July 2018 and January 2019. However, in comparison with the monitoring data during June 2017 and January 2019, the mean values of dissolved heavy metals in water decreased continuously. The mean values of dissolved heavy metals in water monitored during June 2017 and January 2019 decreased on the following order: Zn ≥ Cd ≥ Pb ≥ Ni > Hg.

The mean values of dissolved mercury (Hg) monitored in water during January 2017 and January 2019 were in the range of 0.001 to 0.008 µg/L which were detected at very low concentrations compared to other dissolved heavy metals. The mean values of dissolved cadmium (Cd) and lead (Pb) in water during January 2017 and January 2019 were detected at higher concentrations than dissolved mercury (Hg). The mean values of dissolved lead (Pb) in water in January 2017 were detected at very high concentrations compared to the dissolved cadmium (Cd) and other dissolved heavy metals. However, the mean values of dissolved lead (Pb) in water during June 2017 and January 2019 were detected at lower concentrations than in January 2017. During June 2017 and January 2019, the mean values of dissolved lead (0.002-0.157 µg/L) in water were similar to the dissolved cadmium (0.001-0.192 µg/L), zinc (0.002-0.201 µg/L) and nickel (0.003-0.052 µg/L). The results of this study were similar to the results of Polprasert, 1982 [1] who found that accumulation of cadmium (Cd) and lead (Pb) in water at the Chao Phraya River Mouth was significant.

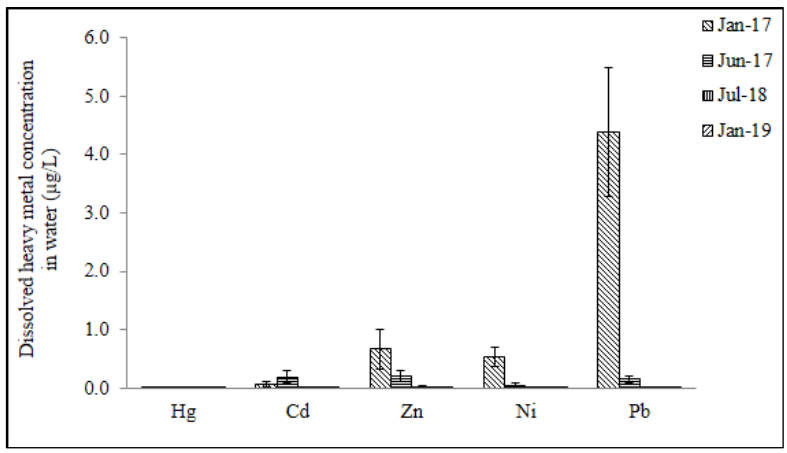

Figure 4 Mean values of dissolved heavy metals in water in the Chao Phraya River Mouth during 2017 and 2019. Each error bar indicates the standard deviation of water samples collected from 20 sample locations of the Chao Phraya River Mouth.

The variation of dissolved heavy metals in the river mouth water might be due to accumulation and release of dissolved heavy metals from the river suspended and bottom sediments. During transportation of heavy metals in the riverine system, it might undergo frequently changes due to dissolution, precipitation and sorption phenomena [6, 8]. According to Huang et al., 2017 [19], heavy metals (Cr, Ni, Cu, Zn, Cd and Pb) in sediment of the Huangpu River, China had a low release flux, among them Cu and Pb had the highest release flux, and dissolved Cu and Pb were fluctuating during the experimental period.

Some heavy metals dissolved in the river water might be attributed from the anthropogenic origins which include discharge of heavy metal-contaminated industrial effluent, either treated or without treatment. However, the heavy metal-contaminated wastewater of factories in the industrial zone, park and estate in cities located along the Chao Phraya River, including Ayutthaya, Phatumthani, Bangkok and Samutprakan is treated before discharge of industrial effluent into canals and rivers, regarding law-established industrial effluent standard, result in the discharge of low amount of heavy metals contained in industrial effluent into the water bodies. According to Mingkhwan and Worakhunpiset, 2018 [20], the levels of heavy metals (Cd, Cr, Cu, Mn, Ni, Pb and Zn) in the surface water collected near the industrial estate in Uthai district and Bangpa-in district, Ayutthaya, as the water source receiving industrial effluent did not exceed the permissible surface water quality standard in Thailand.

Figure 4 shows that the concentrations of heavy metals in water of the Chao Phraya River Mouth in June 2017 and July 2018 in wet season were higher than those concentrations in January 2019 in dry season. This might be due to a period of high rainfall and subsequent fast river flow involving movement of suspended sediment and release of more dissolved heavy metals in the waterway system. According to Mclaren et al., 2004 [3], in August and September (wet season) the change occurred giving rise to the increased cadmium concentrations in water of the Chao Phraya River.

The mean values of all dissolved and total heavy metals (Hg, Cd, Zn, Ni and Pb) in water monitored at the Chao Phraya River Mouth during 2017 and 2019, as shown in Table 2, were lower than the coastal water quality standard in Thailand. The heavy metals in water monitored at the mouths of other World rivers are also shown in **Table 2**. According to Chaiyara et al., 2013 [21], the concentrations of Cd, Zn and Pb in water monitored at the mouths of the Chao Phraya River, the Tha Chin River and the Mae Klong River, Thailand in 2013 were lower than those heavy metals in water monitored at the Chao Phraya River Mouth, Thailand during 2017 and 2019. Compared with the heavy metals in water in the coastal area of other countries, the concentrations of Pb in water monitored at the Chao Phraya River Mouth, Thailand were similar to lead (Pb) in water monitored at the coastal area of Tuaran, Sabah, Malaysia and the Yangtze River Estuary, China [12, 16]. And, the concentrations of Hg and Zn in water monitored at the Yangtze River Estuary, China were higher than those concentrations in water monitored at the Chao Phraya River Mouth, Thailand. In the Odiel River Mouth, Spain, the concentrations of heavy metals in water were very high. The heavy metal-contaminated water in the Odiel River Mouth was caused by acid mine drainage from the abandoned mines and waste in mine dumps [22].

Table 2 Heavy metals in water monitored in the mouths of the Chao Phraya River, Thailand and other World rivers

Metals	Chao Phraya River Mouth Water, Thailand		Chao Phraya River Mouth Water, Thailand	Tha Chin River Mouth Water, Thailand	Mae Klong River Mouth Water, Thailand	Odiel River Mouth Water, Spain	Water of Coastal area of Tuaran, Sabah, Malaysia	Yangtze River Estuary Water, China	Coastal water quality standard in Thailand
	DHM	THM							
Hg, µg/L	0.001-0.008	0.001-0.007						0.036	0.1
Cd, µg/L	0.001-0.192	0.029-0.193	0.010-0.030	0.010-0.030	0.020	100	0.337	0.041	5
Zn, µg/L	0.002-0.668	0.102-1.047	0.150-0.190	0.360-0.530	0.160-0.190		0.822	8.910	50
Ni, µg/L	0.003-0.540	0.052-0.541				300			-
Pb, µg/L	0.002-4.392	0.190-4.428	0.330-0.430	0.150-0.430	0.080	240	5.560	0.829	8.5
Reference	This study		[21]			[22]	[16]	[12]	PCD

DHM: Dissolved heavy metals, THM: Total heavy metals

Inflow of freshwater and input of total heavy metals to the Chao Phraya River Mouth during 2017 and 2019 are shown in **Table 3**. The results indicated that input of total heavy metals to the Chao Phraya River Mouth varied with inflow of freshwater, except in January 2017. In January 2017 (dry season), the Chao Phraya River Mouth received low freshwater inflows, 9.1×10^6 m^3/d, but the contamination of heavy metals entering the river mouth was concentrated – the load per unity of volume, 6.097 µg/L. Consequently, the input of total heavy metals to the river mouth was very high, 55.483 kg/d. In June 2017 and July 2018 (wet season), the Chao Phraya River Mouth received high freshwater inflows, $21.4\text{-}62.9 \times 10^6$ m^3/d, but the contamination of heavy metals entering the river mouth was diluted – the load per unity of volume, 0.657-0.659 µg/L. Consequently, the input of total heavy metals to the river mouth was high, 14.059-41.254 kg/d. In January 2019 (dry season), the Chao Phraya River Mouth received low freshwater inflows, 8.3×10^6 m^3/d, and the contamination of heavy metals entering the river mouth was diluted – the load per unity of volume, 0.667 µg/L. Consequently, the input of total heavy metals to the river mouth was very low, 5.537 kg/d. The variation of total heavy metals to the Chao Phraya River Mouth was observed, most probably related to seasonal river flows and heavy metal contamination levels in water.

Table 3 Inputs of total heavy metals to the Chao Phraya River Mouth during 2017 and 2019

Period of time	Inflow (m^3/d)	Input	Hg	Cd	Zn	Ni	Pb	TML
January 2017	9.1×10^6	µg/L	0.007	0.074	1.047	0.541	4.428	6.097
		kg/d	0.064	0.673	9.528	4.923	40.295	55.483
June 2017	62.6×10^6	µg/L	0.006	0.193	0.218	0.052	0.190	0.659
		kg/d	0.376	12.082	13.647	3.255	11.894	41.254
July 2018	21.4×10^6	µg/L	0.003	0.056	0.177	0.161	0.260	0.657
		kg/d	0.064	1.198	3.788	3.445	5.564	14.059
January 2019	8.3×10^6	µg/L	0.001	0.029	0.102	0.320	0.215	0.667
		kg/d	0.008	0.241	0.847	2.656	1.785	5.537

TML=total metal load

Conclusion

The enriched concentrations of heavy metals (Hg, Cd, Zn, Ni and Pb) were observed in suspended sediment and bottom sediment of the Chao Phraya River Mouth during 2017 and 2019. The observed order of heavy metal concentrations in sediments of the Chao Phraya River Mouth was as follows: Pb > Ni > Zn > Cd > Hg in suspended sediment and Zn > Pb > Ni > Hg > Cd in bottom sediment.

The mean concentrations of all dissolved and total heavy metals (Hg, Cd, Zn, Ni and Pb) in water monitored at the Chao Phraya River Mouth during 2017 and 2019 were lower than the coastal water quality standard in Thailand. The observed order of dissolved heavy metal concentrations in water of the Chao Phraya River Mouth was as follows: Zn ≥ Cd ≥ Pb ≥ Ni > Hg.

The high levels of heavy metal concentrations in suspended and bottom sediments of the Chao Phraya River Mouth might be caused by weathering of rock through water and wind, suspended sediment transport to the river mouth by river flow, tide and seasonal monsoon, and bottom sediment deposition, especially at the inside of river bends and river mouths. Because of the high freshwater inflow in the Chao Phraya River and the discharge of low amount of heavy metals in the treated industrial effluent into the Chao Phraya River, possible sources of heavy metals in suspended and bottom sediments of the Chao Phraya River Mouth were not anthropogenic, but rather lithogenic.

The seasonal variation of heavy metals in suspended and bottom sediments of the Chao Phraya River Mouth was observed, related to the fast river flow in wet season, result in more suspended sediment transported to the river mouth or bottom sediment flushing. The spatial variation of heavy metals in suspended and bottom sediments of the Chao Phraya River was existed, due to adsorption and co-precipitation of dissolved heavy metals onto suspended and bottom sediments and aggregation-sedimentation of suspended sediment during transport.

The seasonal variation of dissolved heavy metals (in µg/L) in water of the Chao Phraya River Mouth was observed, due to accumulation and release of dissolved heavy metals from the river suspended and bottom sediments. The seasonal variation of total heavy metals (in kg/d) to the Chao Phraya River Mouth was observed, related to the river flows and heavy metal contamination levels in water.

Acknowledgement

This study was financially supported by the National Research Council of Thailand (NRCT) and the Department of Environmental Engineering, Faculty of Engineering, Kasetsart University.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 45-54
www.eeat.or.th

Geographic Information System in Managing Flood Protection

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Abstract

Geographic information system (GIS) is a very powerful tool in data preparation, management, manipulation, analysis and presentation. GIS is used in almost all applications. Quantum Geographic Information System or QGIS is free and open-source software. In this study, positions obtained from a mobile mapping system (MMS) were input to QGIS program to demonstrate how to manage flood protection. King Kaeo Road, a southern part of the King's Dyke in the east of Bangkok, was selected as an example for displaying the result of mobile mapping system (MMS) processing and analyzing the causes of inadequate satellite signals to compute the accurate (±5 centimeters) positions. Google map, Google satellite and OpenStreetMap were plug-in to QGIS program. From displayed map, points outside the King's dyke were eliminated and the gaps (of inaccurate positions) could be seen. To fill in small gaps caused by pedestrian overpasses, missing positions could be interpolated or approximated. Repeating MMS data collection at different satellite geometry could remedy the missing positions in some areas. The last recommendation to complete all needed positions along the dyke was to use a ground survey by differential and profile leveling. Using MMS to collect the data and QGIS to help analyzing the result was the most effective and rapid method to create the data base for managing flood protection.

Keywords : GIS; QGIS; Mobile mapping system; flood protection

Introduction

The event of 2011 Thailand flood in Chao Phraya basin caused severe damage to agriculture, industry, economy, society and impact to other regions. This event brought widely management plan for flood protection in the basin. Roads and their barriers were used as flood protecting dykes. Roads were resurfaced frequently, i.e. their heights changed. Wichiencharoen and Santitamnont (2020) [1] proposed to use a mobile mapping system (MMS) for collecting and processing such data, because MMS is the best system to easily and rapidly update the database whenever wanted.

Mobile Mapping System is a system to collect geo-information from mobile vehicles on ground, on water and in the air. System outputs include data for geographic information system (GIS), a digital map, geo-referencing images and videos. A system mainly consists of a Global Navigation Satellite System (GNSS) receiver and an inertial navigation system (INS) for geo-referencing positions of the vehicle [2, 3]. Other equipment on board includes photographic and video cameras, radar, laser, LiDAR and other remote sensing systems [4, 5] . Therefore, data collection and processing using MMS are suitable for flood protection planning because it is easy and fast to obtain heights and can be done at any time needed. The standard equipment installed in MMS is adequate to obtain high accurate heights.

Wichiencharoen and Santitamnont [1] used 3 areas for the study. These were (1) Uta-yarn Avenue in Taweewatana District, approximated 4 kilometers length, (2) Klong 10 to Klong 13 Roads in Patumtani, 30 km length, and (3) the King's Dyke in the east of Bangkok, 60 km length. The King's Dyke is the flood protection dyke of Bangkok. The study result was that it was possible to obtain heights at the accuracy level of ±5 centimeters, which was sufficiently accurate for planning flood protection. Guidelines to obtain heights as many points as possible at the required accuracy level were suggested at the end.

Geographic information system (GIS) is very powerful tool in data preparation, management, manipulation, analysis and presentation [6]. GIS is used in most applications including engineering, planning, management, transport and logistics, insurance, telecommunications, and business. GIS can be viewed as a digital map with details attached to coordinates/locations of interested features on the map.

Quantum Geographic Information System or QGIS is free and open-source software. Free means users do not pay any cost for using the software while they have to pay for commercial software. Open-source means computer coded languages are available so that users, who know the computer language such as C++, Python etc., can add-on or modify the program to their own needs. The first version: 1.0 Kore was posted in 2009. Since then users around the world have helped each other to develop new versions every day. QGIS version 3.10 A Coruna was used in this study, see [7] for a user's manual. Public information such as Google map, Google satellite, OpenStreetMap* can be plug-in to QGIS. Therefore, it is very convenient to the users because all of these data are up to date all the time. Google map and Google satellite are inserted as background of the map while OpenStreetMap can be considered as an attribute of a point.

Aims of this study were (1) a procedure to determine heights at the high accuracy as many points as possible and (2) using QGIS to manage

*OpenStreetMap is a collaborative project to create a free editable map of the world. The geodata underlying the map is considered the primary output of the project. Wikipedia

data base systematically and analyze causes of inability to receive adequate GNSS signals.

Methodology

Research work flow can be classified as follows:

1. MMS data collection on King Kaeo Road (Route # 3256 is one part of the King's Dyke)

King Kaeo Road was selected for the reasons as follows: (a) the road being a southern part of the King's Dyke in the east of Bangkok which is actually used for flood protection planning, (b) surrounding buildings being densest part along the dyke, and (c) having data from Wichiencharoen and Santitamnont [1] for the result comparison.

Recommendation from Wichiencharoen and Santitamnont [1] for data collection was as follows: (a) collected points should be within 20 kilometers from a base station for kinematic surveying, (b) at the beginning of data collection, the vehicle should stop at least one minute so that the initialization of satellite kinematic survey could be completed, (c) then the vehicle smoothly moved at the speed of 30 kilometers per hour (km/h). Data on both directions of the road were collected.

2. MMS data were processed by Inertial Explorer software of NovAtel's Waypoint® Product Group (2014) [8]. Result of the processing included Coordinated Universal Time (UTC), GPS Time, Location and Orientation of the vehicle with their standard deviations and Orientation of the camera. Headings of the said values were shown in **Figure 1**.

3. Data Analysis to get height accuracy and causes of inadequate satellites for positioning. Results shown in 2. was input to Microsoft Excel to compute roughly different positions at every one GPS second so that the speed of the vehicle in each direction could be checked. Then height difference between GNSS height and GNSS incorporated with IMU (GNSS+IMU) height at each GPS second was computed. If the value of the difference was high (e.g. larger than 10 centimeters), the epoch would be considered as no GNSS reception. The filtered positions were input to QGIS program and each position was shown as a dotted point. Either a Google satellite map or a Google street map could be selected as a background of the map. Then we could see where GNSS signal could not receive properly and where positions were not on the King's dyke. Such points were removed from the data base. Causes of improper GNSS receiving would be analyzed and explained. After analysis, recommendation would follow.

4. Conclusions and Discussion. Significant findings and analysis would be concluded.

Work flow of the methodology is shown in **Figure 2**.

UTCTime (HMS)	GPSTime (sec)	Latitude (deg)	Longitude (deg)	H-MSL (m)	H-Ell (m)	SDHoriz (m)	SDHeight (m)	Q	HzSpeed (km/h)	Pitch (deg)
PitchSD (deg)	Roll (deg)	RollSD (deg)	Heading (deg)	HdngSD (deg)	Omega (deg)	Phi (deg)	Kappa (deg)	HdngSep (deg)	PtchSep (deg)	RollSep (deg)

Figure 1 Headings of values resulted from Inertial Explorer software of Waypoint® Products Group

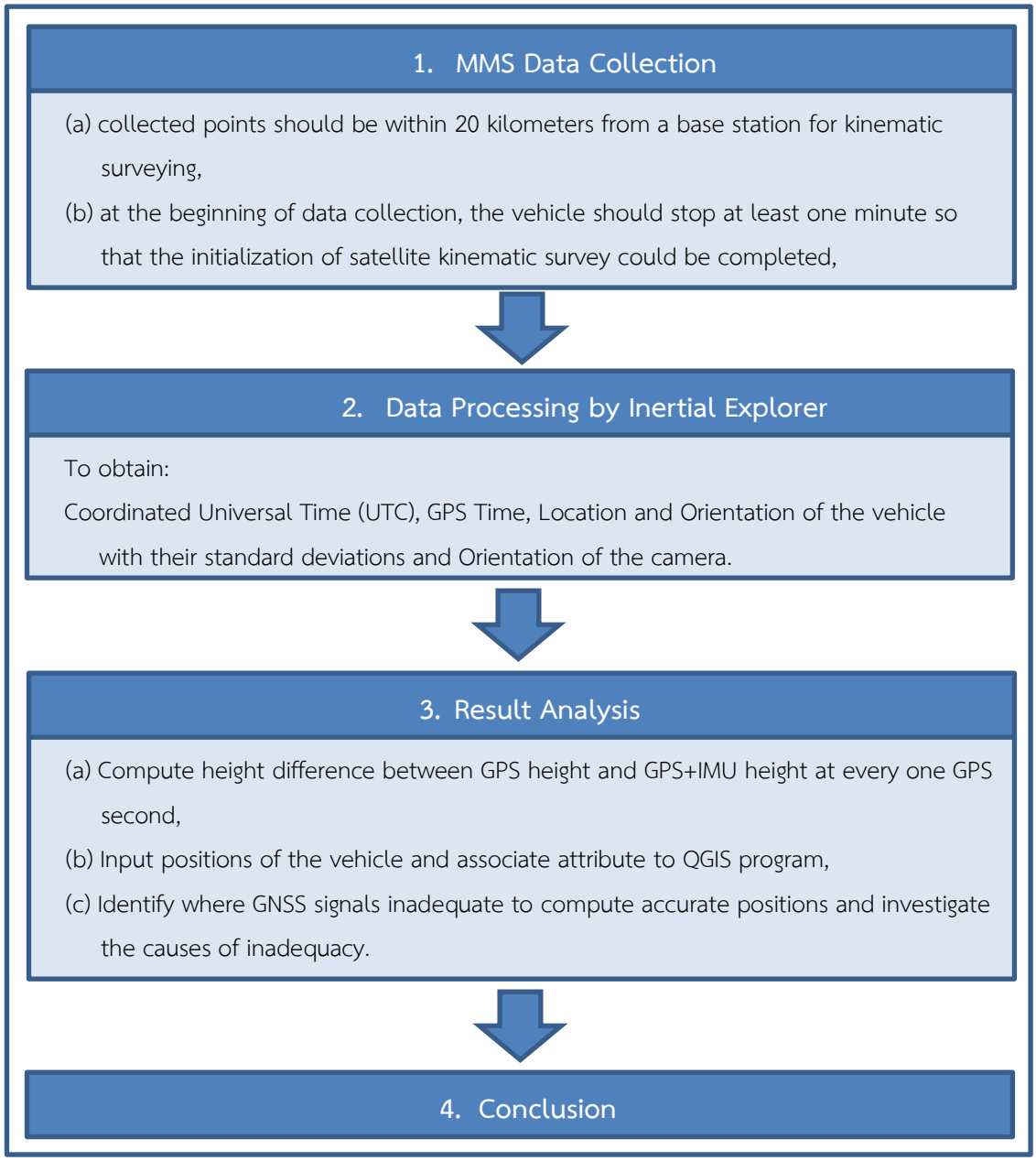

Figure 2 Work flow of the research methodology and step details

Results and Analysis

MMS data along King Kaeo Road were collected on May 21, 2019. The control point SBM.7301/53 of Department of Rural Roads, was used as the base station for the satellite survey. The vehicle speed was kept under 30 km/h. Time to collect of data on the west side of the road was 50 minutes and on the east side was 47 minutes. The route of collection processed at 1-second interval is shown in **Figure 3**.

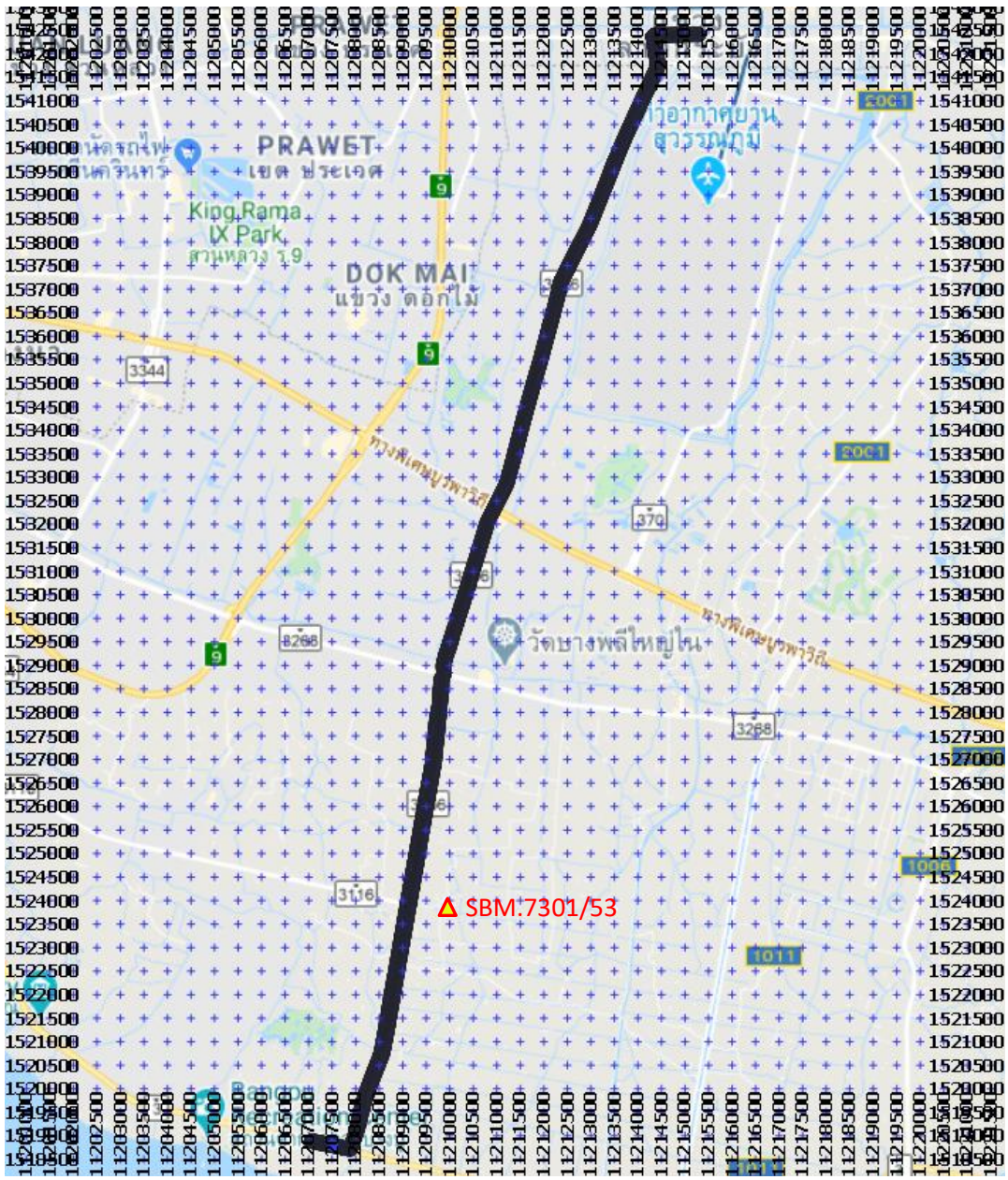

Figure 3 Showing the Route # 3256 where MMS data were collected. The map was plotted by QGIS program and Google Map (accessed on Feb.7, 2020) was plug-in as the background. Grids of 500 meters were shown as crosses

The map in QGIS could be enlarged until individual points were separately seen, e.g. the section shown in **Figure 4**. Missing points (seen as long gaps) were positions where satellite signals were inadequate to compute locations. When Google Satellite was plug-in to QGIS, the cause of signal lost could be seen. On the top of **Figure 4**, the pedestrian overpass appeared and satellite signals were lost just approaching the structure and then needed some seconds to initialize and get back the accurate positions again. The same patterns occurred every time the vehicle approaching the pedestrian overpasses. On the bottom of the figure, Bangna-Chon Buri Expressway could be seen and the same impact to position processing happened as above-said.

Sky plot during the time of data collection on May 21, 2019 can be seen in **Figure 5**. Normally, at least 4 satellites are needed for positioning. At the time of data collection, there were 9 GPS and 6 GLONASS satellites available for positioning. However, under the concrete structures the satellite signals could not pass through.

Figure 4 Showing a section of an enlarged map of **Figure 3**. The red dots were positions of the MMS processing result at every one GPS second. Google Satellite was plug-in as the background

Figure 5 Sky plots at 13.00 hrs. (local time) on May 21, 2019, using [9]. Lines were paths of satellites for the next 6 hours

There was an area that could not receive satellite signals for a long time i.e. when passing Thepharak Road. The vehicle ran along the on-ground road way to serve the purpose of flood protection. While there was an elevated road alongside, this caused inadequate satellites for positioning. There was another area where no overpass structures alongside the road, but the position differences between GNSS and GNSS+IMU were very large. This occurred when the vehicle was moving on the east side and while approaching Sukhumvit Road, see **Figure 6**. Google Satellite map did not show any obstruction around the area. Therefore OpenStreetMap was plug-in to QGIS. Street view was also shown in **Figure 6**. A lot of communication wires between the electric poles could be seen, and these wires obstructed the satellite signals coming from the east side of the sky. The sky plot in **Figure 5** shows that there were four GPS satellites in the eastern sky, so there were five satellites left to compute positions of the vehicles. However, the satellite geometry was not good enough to get accurate positions. This scenario did not happen when the vehicle moving along the west side (northern direction) of King Kaeo Road.

Now the question is what we can do to missing points on the King's dyke. The answer can be classified into three cases.

(a) The first case is points under the pedestrian overpass such as the top section of **Figure 4**. Normally the number of missing points was 5-6, therefore GNSS+IMU heights could be used, see for example **Table 1**. Secondly, linear interpolation between known points could be used. Thirdly, for this particular example, approximate values in centimeter level might be used. Consider the last column of **Table 1**, the heights of the road were almost horizontal, i.e. at 3.80 meters.

(b) The case of **Figure 6**, repeat MMS data collection when satellite geometry is difference or collecting the data at different time of the day, usually 3 hours difference. This choice was confirmed when result of data collected by Wichiencharoen and Santitamnont [1] was used, see **Figure 7**.

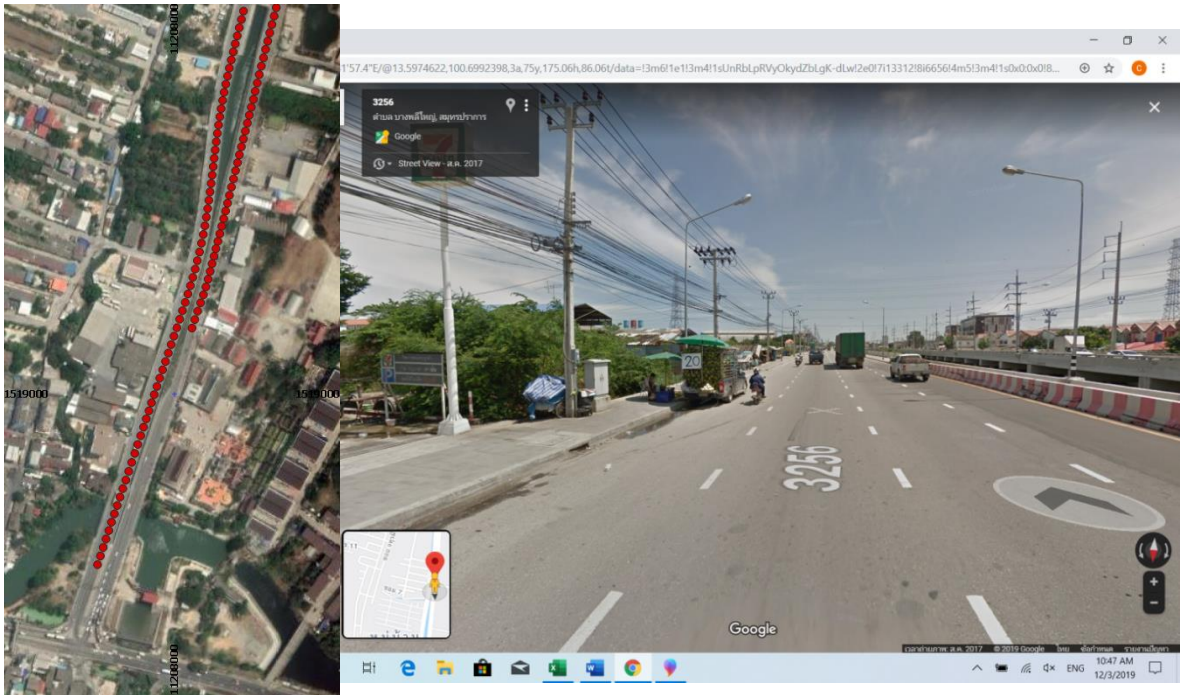

Figure 6 Picture on the left showing Google satellite with accurate positions (red dots) near Sukhumvit road. Moving northward could receive satellite signal better than moving southward. Picture on the right shows the street view (from OpenStreetMap) on the east side of King Kaeo Road (southern direction). A lot of communication wires between the electric poles can be seen. The street view on the opposite side (northern direction), only a few wires can be seen

Table 1 Example of result from Inertial Explorer. MMS data on May 21, 2019 was processed and some selected columns shown

		GNSS only			GNSS+IMU		
Local Time (HMS)	GPS Time (sec)	Latitude (deg)	Longitude (deg)	H-MSL (m)	Latitude (deg)	Longitude (deg)	H-MSL (m)
13:20:38	195656	13.6386	100.7116	3.787	13.6386	100.7116	3.814
13:20:39	195657	13.6386	100.7116	3.815	13.6386	100.7116	3.826
13:20:40	195658	13.6387	100.7117	3.826	13.6387	100.7117	3.821
13:20:41	195659	13.6388	100.7117	3.905	13.6388	100.7117	3.828
13:20:42	195660				13.6389	100.7117	3.789
13:20:43	195661				13.6389	100.7118	3.774
13:20:44	195662				13.6390	100.7118	3.764
13:20:45	195663				13.6391	100.7118	3.809
13:20:46	195664				13.6392	100.7119	3.810
13:20:47	195665	13.6392	100.7119	3.890	13.6392	100.7119	3.798
13:20:48	195666	13.6393	100.7119	3.883	13.6393	100.7119	3.811
13:20:49	195667	13.6394	100.7120	3.758	13.6394	100.7120	3.822
13:20:50	195668	13.6395	100.7120	3.749	13.6395	100.7120	3.804

Figure 7 Showing the same area as Figure 6. The green dots is the positions processed by Wichiencharoen and Santitamnont [1]. Continuous computed positions, when the vehicle was moving southward, can be seen. The result of the old data was different from the new data (in Figure 6)

(c) If the cases (a) and (b) do not work, the only choice left is to use a ground survey. The example of this case is when the vehicle passing Thepharak Road. The known heights of the points nearby can be pinpointed by satellite kinematic survey and used as reference. The method of differential and profile leveling is then performed to get the heights of the missing points, see for example [10]. This method had been used in Wichiencharoen and Santitamnont [1] to verify the accuracy of computed MMS data when satellite signals were insufficient for a long time.

Conclusions and Discussion

This paper demonstrates how to use QGIS in managing flood protection. The new work followed the work of Wichiencharoen and Santitamnont [1]. King Kaeo Road or Highway Route # 3256 (one part of the King's Dyke) was selected for MMS data collection. Inertial Explorer software of NovAtel's Waypoint® Product Group (2014) [8] was used to process the data and obtain the positions of the vehicle. Microsoft's Excel program was used to analyze the accuracy of the points by comparing positions from GNSS only and GNSS+IMU. The result from Excel was then put into QGIS program. The points outside King's dyke and those with inadequate satellite signals were cut off. When the map in QGIS was enlarged until computed points represented by dots were separated, the missing locations would be seen as gaps. The causes of missing were analyzed with the help of plugin Google Map, Google Satellite and OpenStreetMap. The points with

inaccurate locations could be remedied and classified into three cases. First, the heights interpolated by using GNSS+IMU heights, linear interpolation or approximating from the surrounding point. Secondly, repeat data collection in the area when satellite geometry was different from the previous geometry, usually three hours difference. Thirdly, use differential and profile leveling to fill in the gaps, with the help of GNSS kinematic survey.

Now it can be seen that using MMS is the rapid and easy method to collect heights in order to create data base, and using QGIS to effectively manage flood protection planning.

Additional Comment

Photographic and/or video cameras are standard equipment of a mobile mapping system. This means that facilities and other information along the road can be geo-referenced and their locations are included in GIS. AN environmental engineer can use data in the system for wastewater management, air pollution control, etc.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 55-62
www.eeat.or.th

Material Flow Analysis of Lead in Lead Acid Batteries Supply Chain Toward Circular Economy

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Abstract

In Thailand, number of cars using conventional Internal Combustion Engine (ICE) is increasing every year. Due to incomplete combustion by the engine, these cars produce PM 2.5 particles which are dangerous to human health. End of Life (EOL) batteries from these cars, if not properly managed, also cause environmental risks. In this study both primary and secondary data are collected and used for MFA analysis of lead in lead acid batteries. It was found that Thailand produced 17,841,371 batteries (equivalent to 245,140 tons of lead per year) in 2018. Some of these batteries were exported to neighboring countries (equivalent to 82,798 tons of lead per year). Some were sold in country for use in industrial factories and for use in vehicles (equivalent to 5,478 tons and 155,269 tons of lead per year, respectively). The total quantity of lead in battery wastes was 160,747 tons per year. The total quantity of lead recycled by 9 legally registered smelters was only 86,900 tons per year. The remaining 73,847 tons of lead in battery wastes were lost from proper recycling system. The management of this portion of battery wastes remains unknown. Proper recycling by legal smelters can reduce the quantity of lead imported from other countries and decreases the demand for natural resources. The researchers propose the following strategies as a guideline for management of lead in vehicle battery industry toward persistent circular economy. They propose that the government should give support to smelters by offering more tax incentive measures and should support the increasing in their productivity. The government should cooperate with battery manufacturers to nominate representative agency governed by governmental officers to buy EOL batteries from both small and big antique shops. The government should liberalize the investment or invest jointly with private sectors in building lead smelters.

Keywords : Material Flow Analysis (MFA); Lead Acid Batteries; Circular Economy

Introduction

In Thailand, batteries used in most cars are lead acid batteries which are the types of batteries used for vehicles with Conventional Internal Combustion Engine (ICE). Thailand can produce lead acid batteries for their own use and for export to other countries. About 80% of the cars in Thailand use lead acid batteries [1]. This type of batteries has the average life span of 2 to 3 years [2]. After the end of life (EOL), it is considered to be a hazardous waste. The main element from battery wastes that is hazardous is lead. If not properly managed, lead from lead acid battery wastes can contaminate the environment such as water reservoirs, the earth and surrounding air, spreading to human, animals and plants. It can cause environmental and health risks. Lead enters human body through respiration or digestive system. Inside the body it accumulates in the blood, soft tissues, teeth and bones and can cause lead toxicity. It causes adverse effects on central and peripheral nervous system, reproductive organs, kidneys, cardiovascular system and vitamin D metabolism [3]. Also perinatal and neonatal exposures to lead can cause the decrease of neurobehavioral and visual-motoric functions. Lead may also have carcinogenic effect. Recycling is one of the proper methods to manage the lead wastes from EOL batteries. Used batteries are collected and sent to legally registered smelters for proper lead recycling.

However, for some reasons, some of used batteries might be sent to illegal smelters and are not properly managed. At these illegal plants they employ workers to cut the batteries using axes and smelt the batteries together with their plastic boxes and lead grids in a reverberatory furnace without any protective equipment. The recovery efficiency of this technology is very low with high emission of pollutants. It is dangerous to employees and the environment as well [4]. Proper battery recycling not only reduces the amount of wastes, but also decreases the demand for natural resources.

Material flow analysis is a systematic assessment of the flows and stocks of materials within a system defined in space and time. It is an invaluable tool used in resource management, waste management and environmental management. It was postulated by Greek philosophers more than 200 years ago. After that, around 40 years back, Abel Wolman introduced the term “metabolism of cities” which coined the city as a living organism with inputs, stocks and outputs of material and energy [5]. MFA is a tool to analyze the metabolism of materials in order to analyze material flows and stocks within a given system [6]. It can be applied to Evaluate the importance and relevance of the flows and stocks ,Control material flows and stocks to support certain goals such as sustainable development, Assess resource utilization and environmental impacts, Set up long term environmental policy and resource management strategy and Understand and control material flow of heavy metal containing batteries.

In the management system for circular economy, system must be viewed holistically, at the beginning, the middle and the end of material flow to get the highest cyclical flow (life cycle) starting from the utilization of raw materials for battery production. This study applied Material Flow Analysis (MFA) as a tool to trace lead flow and stock in lead acid batteries supply chain in Thailand in order to analyze the current status of lead management and the way for improvement towards circular economy.

Methodology

This study applied Material Flow Analysis (MFA) as a tool used for systematic assessment of the flows and stocks of materials in each unit and the whole system within a defined temporal and spatial system [7]. The research methodology of this study could be described in 4 steps as follow:

1) Setting the scopes of the analysis study, identifying the system boundary and components

The target and the boundaries of time and space were decided. The target in this study was to establish the material flow system of lead in lead acid battery. The time or temporal boundary was the year 2018 and the spatial boundary was Thailand. The life cycle chain of lead in lead acid battery consisted of three stages, as shown in **Figure 1**, including product manufacturing, product use and waste management. It should be noted that there was no lead mining in this life cycle chain. In Thailand there has been no lead mining since 2001 [8], because of environmental problems, such as contamination of stream water by lead ore tailings, impacts on health and well being of people in the local area. Lead which is a raw material for battery production came from import and recycling only. The life cycle chain of lead in lead acid battery in Thailand was different from other studies in that there was no resource mining. Most other studies included resource mining as a stage of the life cycle chain.

2) Data acquisition (Data collection)

Both primary and secondary data were collected.

(1) The secondary data were collected by searching the official websites of governmental offices such as the Department of Primary Industries and Mines, the Department of Land Transport and the Office of Industrial Economics, and the articles published in local language or Thai. The data about the quantity of lead and lead acid battery in Thailand were gathered from the official websites of governmental offices.

(2) The primary data were collected by interviewing the customers or battery users and the battery selling shops in Chiang Mai.

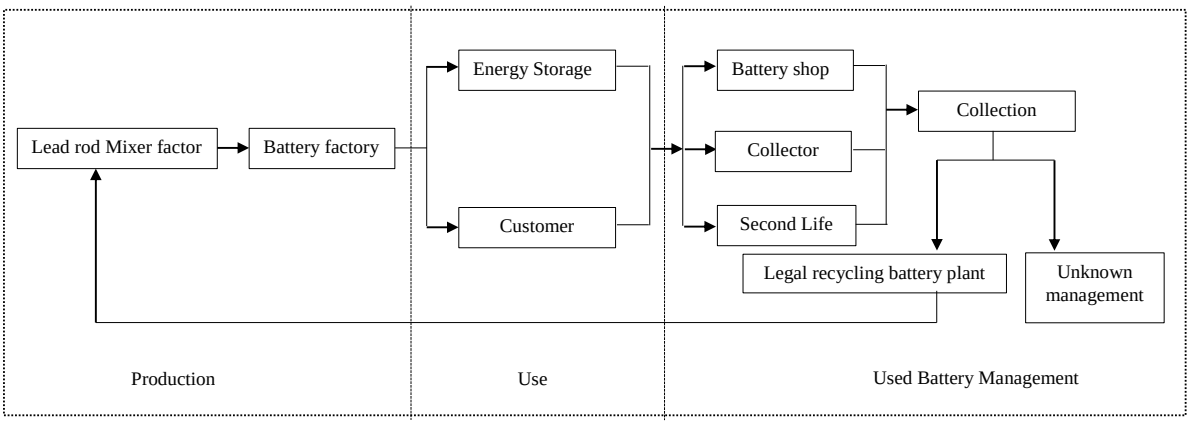

Figure 1 Scope of MFA of lead in Lead Acid Batteries

3) Schematic modeling and balance for material flow system framework

This step constructs the system to use the data collected above. When some data was not acquired, the mass balance or so-called mass conservation, i.e. mass-in is equal to mass-out can be used to balance the materials. Software STAN was used to do Material Flow Analysis of the real current situation.

4) Interpreting MFA result for lead in lead acid battery

In this step, the results of the above MFA for lead in lead acid battery were interpreted, to find out the appropriate method for battery waste management system in Thai.

Results and Discussions

It was shown in this **table 1** that the total number of lead acid batteries produced in Thailand in the year 2018, was 17,841,371. Out of this number, 11,300,511 were sold in country for use in vehicles (cars and motorcycles), 398,711 for use in industrial factories and 6,026,025 were exported to other countries. (**Figure 2** shows the diagram of the material flow of lead in lead acid battery via the MFA method)

Table 1 The quantity of lead in lead acid battery in each process

No.	Process	No. of battery per year	Estimated quantity of lead (Kg prt year)*	Quantity of lead (Tons per year)	Source
1	Import lead	-	-	161,050	Department of Primary Industries and Mines 2018
2	Export Battery	6,026,025	82,797,584	82,798	Office of Industrial Economics 2018
3	Lead rod /mixer factory	-	-	247,950	Import Lead-Legal recycle
4	Battery factory	17,841,371	245,140,438	245,140	Office of Industrial Economics 2018
4.1	Energy storage	398,711	5,478,289	5,478	Batteries sold in country (11,699,222) – No. transportation vehicles (11,300,511) = 398,711
4.2	Customer	11,300,511	155,269,021	155,269	Department of Land Transport 2018
4.2.1	Battery shop	8,136,368	111,793,695	111,794	72% of customer (from interview data)
4.2.2	Collector	1,695,077	23,290,353	23,290	15% of customer (from interview data)
4.2.3	Second life	1,469,066	20,184,973	20,185	13% of customer (from interview data
5	Collection	-	-	160,747	Energy storage +Battery shop +Second life + Collector
5.1	Legal recycling battery plant	-	-	86,900	Article on material flow of lead in lead acid battery 2017
5.2	Unknown management	-	-	73,847	Collection-Legal recycling

*estimation was based on “quantity of lead in lead acid battery =13.74 kg/battery

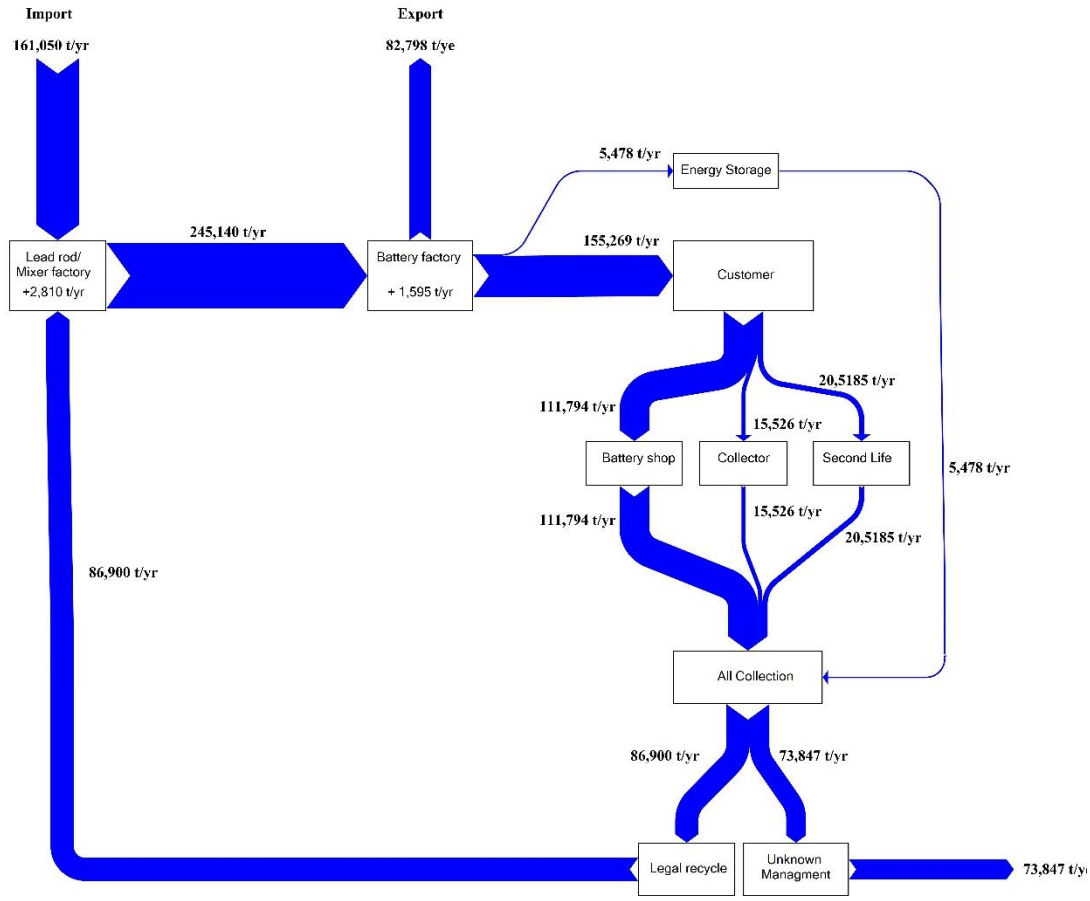

Figure 2 Diagram showing the material flow of lead in lead acid battery by MFA method

The overall input of lead in the year 2018 was 161,050 tons. It was found that in Thailand there has been no lead mining in Thailand since 2001 [8], because of environmental problems, such as contamination of stream water by lead ore tailings, impacts on health and well being of people in the local area. Therefore, lead which is a raw material for battery production came from import and recycling. This amount of lead input was totally imported in the form of lead rod from other countries. The imported and the recycled lead were, after that, used in the production of lead acid batteries. The total amount of lead used in battery production was 245,140 tons [9]. Data from the Department of Land Transport 2018 showed that The amount of lead in lead acid batteries that were exported to neighboring countries was 82,798 tons. The amount of lead in batteries that were sold in country for use in industrial factories and for use in vehicles was 5,478 and 155,269 tons respectively.

The lead acid batteries that have been used for a long time and lost their capacity to the point when no longer suitable for use in vehicles.The primary data from interviewing with the customers showed that among the old (EOL) batteries 72% of them were traded in for new ones in battery shops, 15% were sold to antique shops for recycling and 13% were reused as stationary storage for photovoltaic (PV) energy in second life application. Using these percentages

of EOL batteries for calculation, it was found that the amount of lead in lead acid batteries which were traded in for new ones in battery shops, sold to waste collection and recycling shop and reused in second life application were 111,794 tons, 23,290 tons and 20,185 tons respectively.

From interviewing with the battery selling shops, it was found that about 30% of the old EOL batteries that they bought were resold to antique shops, and 50% were resold to the company shops bigger than the antique shops. There was no information about how the remaining 20% of EOL batteries were managed.

The old EOL batteries collected at battery selling shops, small antique shops and bigger company antique shops eventually were sent to smelters for recycling. Thailand has only 9 lead smelters that are legally registered [10]. The quantity of lead recycled by these smelters was only 86,900 tons per year. Compared with the lead demand of the country, the quantity of lead recovered from recycling process is still not adequate. To get adequate quantity of lead for the production of new batteries, 161,050 tons of lead was imported.

The total quantity of lead in lead acid battery wastes was 160,747 tons per year. The quantity of lead recycled by the legally registered smelters was only 86,900 tons per year. It is not known how the remaining 73,847 tons per year of lead in battery wastes were managed. This amount of lead in battery wastes were not in the legally registered recycling system. It could be assumed that some portion of this amount of lead was sold for battery recycling, to lead smelters which are not registered to the Department of Industrial Works. This is because the illegal or unregistered smelters paid a higher price to the EOL batteries than the legally registered smelters. The other portion might be sold to unregistered smelters for production of lead rods. These lead rods would be sold to factories making ball bearings, trawl, seine, fishnet etc. Illegal smelting that does not meet the standard can cause danger. People will have the chance to directly expose to lead by inhalation or direct contact. Prolonged and continued exposure to lead can cause lead toxicity. It can cause death if the body suddenly receives a large quantity of lead. People may receive lead indirectly by drinking water or eat meat and vegetables contaminated with lead etc.

Waste and Hazardous Substances Management Bureau, Pollution Control Department [11] states that EOL batteries are considered to be hazardous wastes from the community. Forty per cent (40%) of the battery wastes were managed properly. Sixty per cent (60%) of them were not properly managed. The main problem is that the garbage dumping place cannot prevent the dissemination of hazardous wastes. The hazardous wastes are thrown away mixing with general wastes. The Waste and Hazardous Substances Management Bureau, Pollution Control Department has therefore the policy to manage the hazardous wastes following the environment control management plan as follows by Reduce the quantity of wastes by the 3R principle, namely reduce, reuse, recycle and encourage the use of materials that are friendly to environment, Have the system to bring back hazardous wastes from used products and Promote the investment of private sector to build smelter or the center for proper discarding hazardous wastes.

The quantity of lead imported from other countries would be reduced if the 73,847 tons of lead in battery wastes were brought into an appropriate or proper recycling system, making

the system move closer to circular economy. Nevertheless, there must be a thorough study on how to collect and send the EOL lead acid batteries into a correct management system.

From this study, the problem in management was found to be at the step of collection of EOL batteries before sending them to smelters. Battery shops sold EOL batteries to small antique shops, and the small antique shops resold them to bigger antique shops. After that it was not clear how they were managed. They could be sold to legally registered smelters or to unregistered smelters. It is not known how many of them were sold to legally registered smelters and how many to unregistered smelters.

The researchers propose the following strategies to manage lead acid battery wastes. They propose that the government should give support to smelters by offering more tax incentive measures and should support the increasing in productivity of the smelters. The government should also cooperate with battery manufacturers to nominate representative agency governed by governmental officers to buy EOL batteries from both small and big waste collection and recycling shop. The government should also liberalize the investment of private sectors or make joint investment with them in building lead smelters.

Conclusion

In the year 2018, Thailand could produce totally 17,841,371 batteries. Lead (in the form of lead rod) which is the raw material for battery production came from import (from other countries) and from recycling (in country). Lead acid batteries produced in Thailand were exported to neighboring countries, used in industrial factories and vehicles in country. About 73,847 tons of lead in battery wastes were lost from the system of collection and sending to legally registered smelters. It is possible that this amount of lead in battery wastes were sent to the illegal or unregistered smelters that did not meet the standard and could cause bad effects or problems to the environment. The quantity of lead imported from other countries would be reduced if the 73,847 tones of battery wastes were brought into an appropriate or proper recycling system, making the system move closer to circular economy. Nevertheless, there must be a thorough study on how to collect and send the EOL lead acid batteries into a correct management system. The proposed strategic plan for EOL battery managements are:

(1) The government gives supports to smelters by offering more tax incentive measures.

(2) Support the Increase in productivity of smelters.

(3) The government cooperates with battery manufacturers to nominate representative agency governed by governmental officers in each province to buy EOL batteries from both small and big antique shops.

(4) The government liberalizes the investment or invests jointly with the private sectors in building lead smelters.

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Thai Environmental Engineering Journal Vol. 34 No. 3 (2020) : 63-70
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Biodegradation of PAHs by The Mixed Cultures of Diesel Degradation Bacteria

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Abstract

Polycyclic aromatic hydrocarbons or polyaromatic hydrocarbons (PAHs) is an organic compound in hydrocarbon group. This structure is two or more aromatic rings without heteroatoms. The most abundant groups of aromatic compounds occurring in diesel fuels are naphthalene. Three strains of diesel-degrading bacteria including *Achromobacter insolitus, Candida spp and Xanthobactor polyaromatici yorans* was proved as high capability to degrade diesel. Thus, this study was aims to determine the optimal condition for growth up of diesel-degrading bacteria and diesel degradation efficiency. The experiment was conduct in batch experiment with the varied ratio of synthetic wastewater as nutrient and diesel concentration with surfactants (N:D) at 100:0, 80:20, 60:40, 40:60, 20:80 and 0:100. Then the optimal ratio of N:D was used to determine the effect of initial naphthalene concentration on naphthalene degradation. The different initial concentration of naphthalene was varied in the range of 0 to 100 mg/L. The results showed that the highest percent COD removal (100%) was found at N:D ratio 60:40 and 0:100 followed by N:D ratio 40:60 at 96.7% and N:D ratio 80:20 and 20:80 at 97.4%, respectively. The different of nutrient added was affected the growth of biomass. The highest biomass yield was found in N:D ratio 20:80. The growth of biomass depended not only diesel but also glucose in nutrient. Glucose play as a cometabolism for growth up bacteria. The initial concentration naphthalene was affected the growing of biomass and the efficiency of naphthalene degradation. The highest naphthalene degradation efficiency (99.8%) was found at initial napthalene concentration at 20 mg/L.

Keywords : Biodegradation; Diesel degrading bacteria; Diesel; Naphthalene

Introduction

Sixteen PAHs are regulated by the U.S. Environmental Protection Agency (USEPA) based on their potential human and ecological health effects including Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz[a]anthracene, Chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a] pyrene, Dibenz[a,h]anthracene, Benzo[ghi]perylene, Indeno[1,2,3-cd]pyrene [1]. Wattayakorn (2012) [2] reported that water sample from the estuary of Chaopraya contained 4.71 µg/g of PAHs, the highest types of PAHS was naphthalene. Naphthalene or mothballs are toxic to human and animals include cause of cancer, tumor, acute toxic of nerves system, hematoma system, respiration system and digestion system.

There are many aspects of relation between diesel and naphthalene. PAHs was found in burning of coal, fuel and part of diesel. Diesel is complex and several structure for example n-alkalines, isoand cycloalkanes, PAHs, sulfer and aromatic compound especially naphthalene and alkylnapthalene [3]. Moreover, in the middle distillates such as diesel fuel is identified by a variety of straight, branched, and cyclic alkanes, as well as naphthalene, methylnaphthalenes [4].

From previous study by Singhyakaew (2015) [5], Three strains of diesel-degrading bacteria were collected from the activated sludge process of An-ping wastewater treatment plant in Taiwan. These cultures are high capability to degrade diesel. Moreover, the single culture of microbe is low capacity than mix culture of bacteria because metabolize a limited scope of hydrocarbon substrates [6]. So, this study was utilized Achromobacter insolitus, Candida spp and Xanthobactor polyaromatici yorans to degrade diesel and naphthalene with cometabolism in metabolic process.

The process of bioremediation was influenced by some physical factors as an example temperature, pH, oxygen, nutrient, microorganism number, consortium of microorganism, bioavailability, contaminant characteristics and toxic of end products [7]. The development of limiting factors can lead to more capacity treatment technology. One important factor to limit degradation of diesel is the lack of carbon source. Nutrient was found to stimulate growth of the pollutant degrading microorganisms and enhance their ability to degrade contaminants [8]. Thus, the aim of this work was investigating the optimal nutrient and diesel ratio to enhance diesel degradation and growth up of microorganism. In addition, the effect of initial naphthalene concentration to naphthalene degradation was investigated.

Methodology

Synthetic wastewater

The synthetic wastewater was prepared based on the study of Banu et al., (2009) [9]. About 20,000 ml of synthetic domestic wastewater was prepared by using various chemical as glucose 4.5 g, NH_4Cl 4g, $NaHCO_3$ 8g, KH_2PO_4 0.5 g, microelement solution ($MnCl_2.4H_2O$ 2 ml, $ZnCl_2.2H_2O$ 2 ml, $CuCl_2.2H_2O$ 2 ml, $MgSO_4.7H_2O$ 2 ml, $FeCl_3.6H_2O$ 2 ml, $CaCl_2.2H_2O$ 2 ml). After that added the RO water until the final volume was 20,000 ml. The synthetic wastewater was used as nutrient for all experiment. Synthetic wastewater was kept in refrigerator (4°c) until used and prepared a new one every 2 days.

Diesel solution

Diesel solution was prepared by getting the pure diesel 10 ml and mixed with 1 mL of Ethylene glycol mono-butyl ether (EGBE), ($CH2OHC_{H2}O_{C4}H9$, 99%) and diluted with reverse osmosis water to obtained the final volume at

1,000 ml. It was stirrer for 15 minutes for dissolve the diesel in water. At last, the 180 ml of diesel solution was diluted again with reverse osmosis water to obtained the final volume at 1,000 ml. The concentration of Diesel is 1.8 $x10^{-2}$ (v/v) and surfactant is 1.8 $x10^{-4}$(v/v). The diesel solution was prepared every two days and keep in refrigerator 4°c.

Mixed culture of bacteria

The mixed culture bacteria were collected from previous study of Singhyakaew (2015) [5] and microbial was cultured for 1 month and was stored for 1-2 years. The mixed culture bacteria consist of Achromobacter insolitus, Xanthobactor polyaromatici, and Candida spp. Those microbes were grown-up by feeding diesel with ethylene glycol mono-butyl ether (EGBT).

Experiment for determine the optimum ratio of synthetic solution (N) and diesel with surfactant (D) for diesel degradation

A batch experiment was conducted with varied ratio of N:D in the range of 100:0 to 0:100. The experiment was conducted with 1,000 ml working volume and start-up with 30 ml of mixed culture bacteria. The experiment was done in aeration mode with controlled DO concentration above 2 mg/L at room temperature. Water samples from reactor was collected and analyzed for their total COD (TCOD) and soluble COD (SCOD) every day. In addition, the mixed liquor volatile suspended solids (MLVSS) was investigated to study the accumulation mixed culture of diesel degradable bacteria. The experiment was operated until the COD concentration was increasing from the previous operating day. Furthermore, various parameters including pH, temperature, DO, and MLSS was also measured every day.

Experiment for determine the effect of initial naphthalene concentration on degradation

Two-liter volume beaker was used as reactor by added the optimal ratio of N:D from the previous experiment. About 30 ml of mixed culture bacteria was added as seed sludge. The different initial concentration of naphthalene which varied in the range of 0 to 100 mg/L. The experiment was done in aeration mode with DO concentration above 2 mg/L at room temperature. Various parameters including pH, oxidation-reduction potential (ORP), temperature, MLSS and MLVSS was measured every day. While naphthalene concentration was measured every 2 hours by HPLC technique with fluorescence detection at 254 nm, the mobile phase was a mixture of water and acetroniitrite (70:30 v/v). Separation was carried out with a reverse phase 5 µm c-18 column (250 x 4.6 mm) with flow rate 1 m/m.

Results and Discussions

The optimal ratio of N:D for diesel degradation

The results in **Table 1** showed that the highest percent SCOD removal was found at batch C and F (100%) followed by batch D (96.7%) and batch B, E (97.4%), respectively. The obtained results showed that the mixed culture bacteria used in this study was high efficiency to remove COD from wastewater. Thus, it can be indicated that the ratio of N:D did not affect the degradation of COD by mixed culture bacteria but affect the degradation time. The higher initial SCOD concentration required more degradation time to degrade SCOD.

Table 1 Percent SCOD removal and degradation time

Batch (N:D)	Initial SCOD (mg/L)	Minimum SCOD (mg/L)	% SCOD removal	Time (hr)
A (100:0)	247	10	96.0%	24
B (80:20)	312	8	97.4%	24
C (60:40)	344	0	100%	24
D (40:60)	390	13	96.7%	24
E (20:80)	464	12	97.4%	48
F (0:100)	509	0	100%	72

The results of pH are shown in **Figure 1**. It was found that pH values in all conditions was in the range of 7.0-8.41 except in batch F which lower than 7. The pH of batch F was decreasing along the experiment period because the stationary phase of batch F was reached earlier than the others batch.

MLVSS was measured to identity the increasing of biomass and determine the growth up efficiency. The results are shown in **Figure 2**. Thus, the results showed MLVSS of all batch was increase in same trend. At start of experiment, the initial bacteria 30 ml was added to reactor. The initial MLVSS in all batch was in the range of 30-47.5 mg/L. After 24 hours, the MLVSS of all batch was increasing. The highest increasing of MLVSS was found in batch E (N:D = 20:80). On the other hands, the lowest increasing of MLVSS was found in batch F (N:D = 0:100). As the increasing of MLVSS represented the growth of bacteria. It can be indicated that the bacteria growth of batch F was lowest. The results were the same with batch A (N:D = 100:0) which found the low growth of bacteria. Thus, it can be concluded that only diesel and only synthetic solution has low ability to growth up bacteria. The growth up of bacteria need both diesel and nutrient in synthetic solution. The optimal ratio of N:D was 20:80 as in batch E that provided the highest increasing of bacteria resulted in high growth rate of bacteria.

The biomass yield was calculated as shown in **Table 2**. This study was focus on the volume of the microbial community because it was necessary for startup the system. The highest biomass yield was found in batch E. The result was same trends as MLVSS results. It indicated that the carbon source in batch E was highly rated to degrade and promote growth up of mix culture. It can be indicated that the synthetic solution added to reactor was affected the biomass growth and the ratio 20:80 of synthetic solution: diesel is the optimum for growth up mix culture to degraded diesel. Thus, the growth of biomass depended not only diesel but also glucose in synthetic solution. Glucose play role in a cometabolism or substrate as was found in the study.

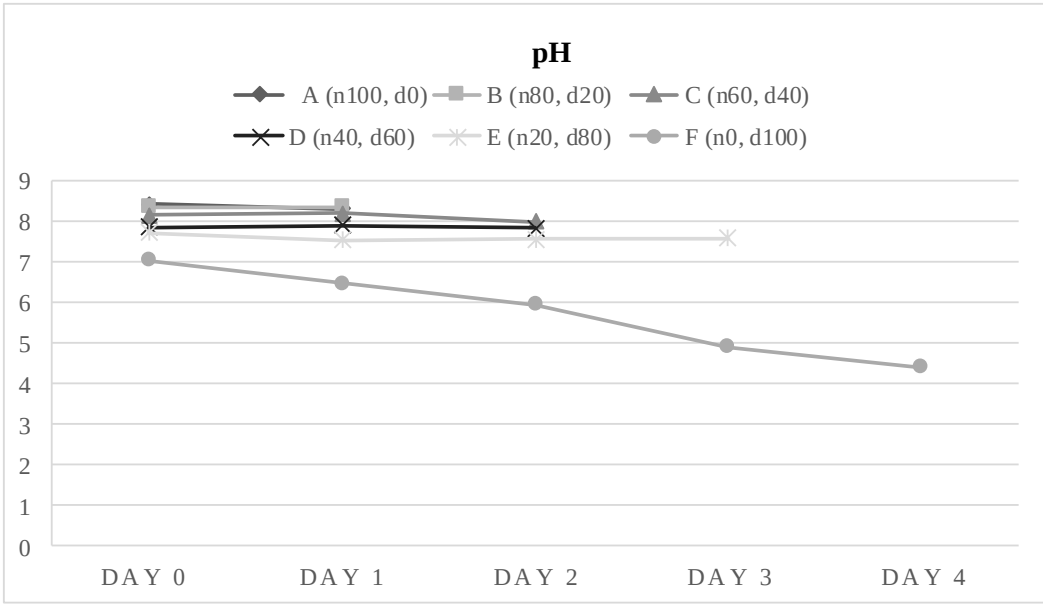

Figure 1 The results of pH in each condition

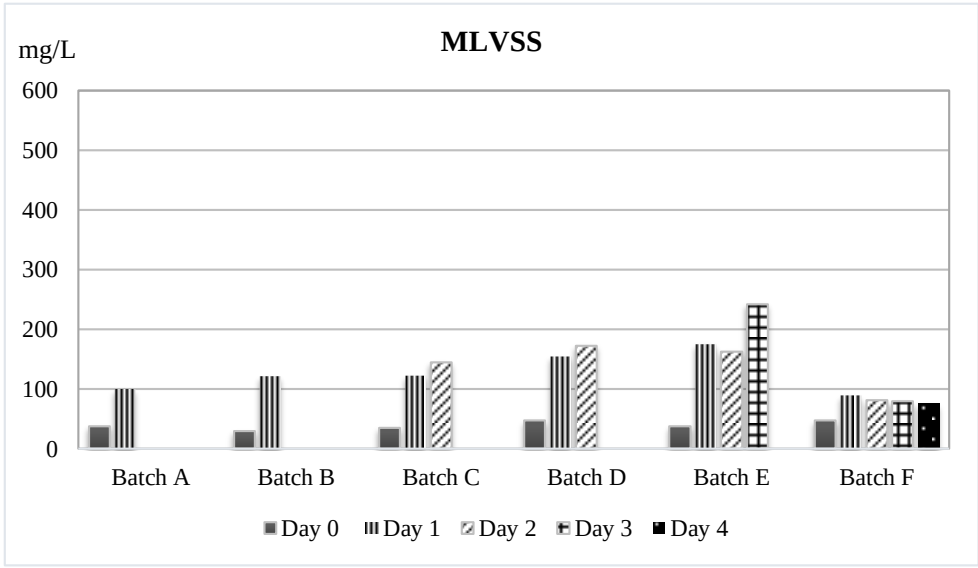

Figure 2 MLVSS in each condition

Table 2 Biomass yield in each batch

	Batch (N:D)					
	A (10:0)	B (80:20)	C (60:40)	D (40:60)	E (20:80)	F (0:10)
Yield (mg of MLVSS/mg of SCOD)	0.2637	0.3026	0.3197	0.3315	0.4535	0.0540

Effect of initial concentration naphthalene on degradation efficiency

The results showed that pH of all batch was found in the range of 6.52-7.99 at all batch conditions**.** The MLVSS in every batch was increase in the same trend. That was indicated varied naphthalene concentration was not affect to growth up the mix culture of bacteria. The biomass yield was calculated to indicate the biomass growth in each batch by normalized the COD concentration in each batch. The results of biomass yield are shown in **Table 3**, it can be indicated that the concentration of naphthalene affects biomass yield. Thus, the optimal initial concentration of naphthalene was 20 mg/L which provided the high growth rate of microbe.

The degradation of naphthalene in each batch was investigated. **Figure 3** showed naphthalene concentration in each batch. The results of degradation of naphthalene was the same trend in all batch. The naphthalene was rapidly degraded in 2 hours and continuous degraded slowly. The highest naphthalene degradation was found in batch A (99.8%) and followed by batch B (87.4%). The naphthalene degradation in batch C to E was the nearly the same (75%). The efficiency and time to degrade naphthalene was affected by the initial naphthalene concentration. The higher decrease naphthalene concentration provided the lower efficiency and required more time to degrade. This resulted can imply that the suitable of initial napthalene concentration was 20 mg/L.

As the government gazette of Thailand, naphthalene does not over 48 mg/L in underground water [10]. Microbe can degrade naphthalene of every batch to lower than 48 mg/L after 2 hours operation periods.

Table 3 Biomass Yield of each batch

	Batch				
	A	B	C	D	E
Initial Naphthalene concentration	20 mg/L	40 mg/L	60 mg/L	80 mg/L	100 mg/L
Yield (mg of MLVSS/mg of SCOD)	1.6	0.5	0.325	0.275	0.195

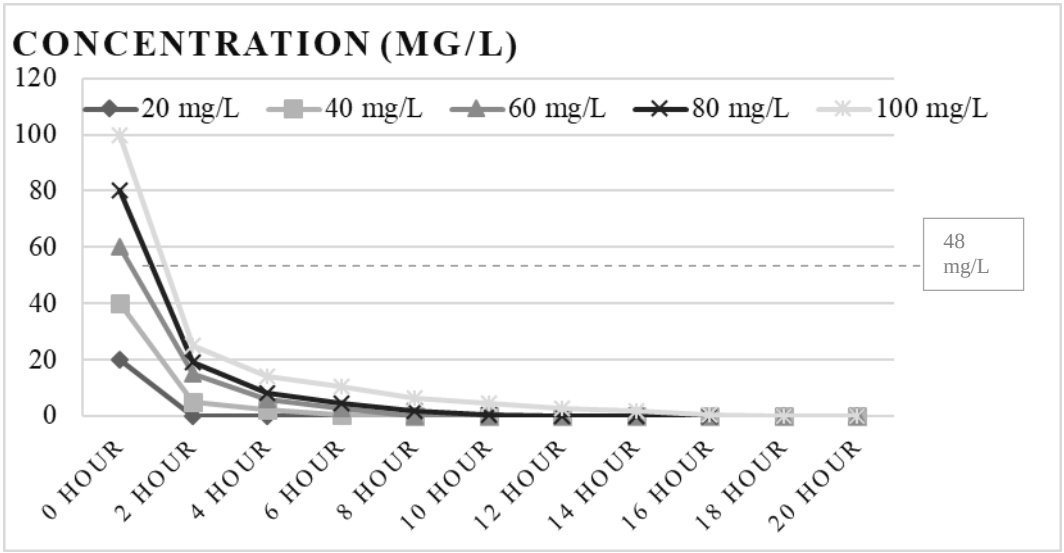

Figure 3 Naphthalene concentration in each batch during operating periods

Conclusion

The mixed culture bacteria was high efficiency to remove organic compound from wastewater. The ratio of N:D did not affect the degradation of COD by mixed culture bacteria but affect the degradation time. The result of MLSS and MLVSS showed that the mix cultured was low degradable when degrade pure diesel or pure synthetic solution. Thus, the ratio of synthetic solution and diesel added was affected the biomass growth. The highest biomass yield (0.4535 mg of MLVSS/mg of SCOD) was found at N:D ratio of 20:80. From the obtained results, the optimal ratio of N:D was 20:80 with the high SCOD removal efficiency and biomass yield.

The mixed cultures of diesel degradation bacteria can degrade naphthalene 100% with varied concentration in varied duration time. The highest efficiency of naphthalene degraded was found in lowest initial concentration of naphthalene (20 mg/L). The suitable initial concentration of naphthalene was 20 mg/L which provided the highest growth rate of microbes and the group of cultures can degrade the others chemical that similar diesel structure as naphthalene.

Acknowledgement

This research work was partially supported by Graduate School of Chiang Mai University and Chia Nan University

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[1] Inthorn, D., Sidtitoon, N., Silapanuntakul, S. and Incharoensakdi, A. 2002. Sorption of mercury, cadmium and lead in aqueous solution by the use of microalgae. Science Asia. 28 (3): 253-261.

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THAI
ENVIRONMENTAL ENGINEERING JOURNAL
Environmental Engineering Association of Thailand (EEAT)

ISSN 1686 - 2961
ISSN 2673 - 0359 (Online)

Vol. 34 No. 3 September – December 2020