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Petroleum Hydrocarbon Pollution in Soil and Surface Water by Public Oil Fields in Wonocolo Sub-district, Indonesia

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ABSTRACT

Public crude oil fields in Wonocolo sub-district were active from 1942 until now and have inadequately operated. The aims of this research were to measure the level of total petroleum hydrocarbon (TPH) pollution and their distribution in soil and surface water at the Wonocolo public crude oil fields. Twelve composite soil samples were collected from uncontaminated and contaminated sites of old well (OW), transportation line (T), and refinery area (R) at the depths of 0–30 cm, 30–60 cm, and 60–90 cm. The composite surface water sample was obtained from two points with different distances from the river side. TPH from soil and surface water samples were extracted using soxhlet and gravimetric method. Quantification of TPH was performed using Fourier Transform Infrared (FT-IR) Spectrometer. From the results of this study, it was concluded that soils and surface water are contaminated by TPH of 119.80–107,190 µg/g and 211,025.73 µg/L, respectively. TPH is clearly located in the upper of 0–30 cm depth at OW, T, and R sites (52,328.14–107,189.63 µg/g). These concentrations exceeded the soil quality standard of TPH and classified as category A for human hazard risk. The findings from this study show that there are considerable health risks which are potentially poisonous to humans in the local area. We recommend that remediation could be conducted using biological methods to reduce TPH pollution level.

Keywords: human risk, soil pollution, surface water contamination, total petroleum hydrocarbon, public crude oil fields.

INTRODUCTION

Public crude oil fields were found in Indonesia. The largest one is located in Wonocolo sub-district, Bojonegoro. In Wonocolo, public crude oil fields area is 11.37 km^2 and it has been managed by local community that utilize the old wells of Dutch colonial period [Marwoto 2010, Naumi and Trilaksana 2015]. Some researchers reported that there were 35–44 active old wells which produced 25.77–50,00 m^3/day of crude oil [Handrianto et al. 2012].

Crude oil was discovered by Adrian Stoop in 1893 at Wonocolo sub-district. It became the first crude oil exploration and exploitation point of Dutch Government [Naumi and Trilaksana 2015]. The oil drilling point at that time is shown in Figure 1. In 1942, the exploration activities at Wonocolo sub-district were taken over by the Japanese Government. After Indonesia gained independence in 1945, the management of the mining activities was transferred to the National Oil Mining Company from 1948 to 1987. Afterwards, the management was handed over to exploration and production unit (UEP) at Pertamina III according to Mining and Energy Ministry Decree No. 0177. Since then, Pertamina UEP III has collaborated with the local community to manage the exploitation activities in Wonocolo public oil fields [Marwoto 2010].

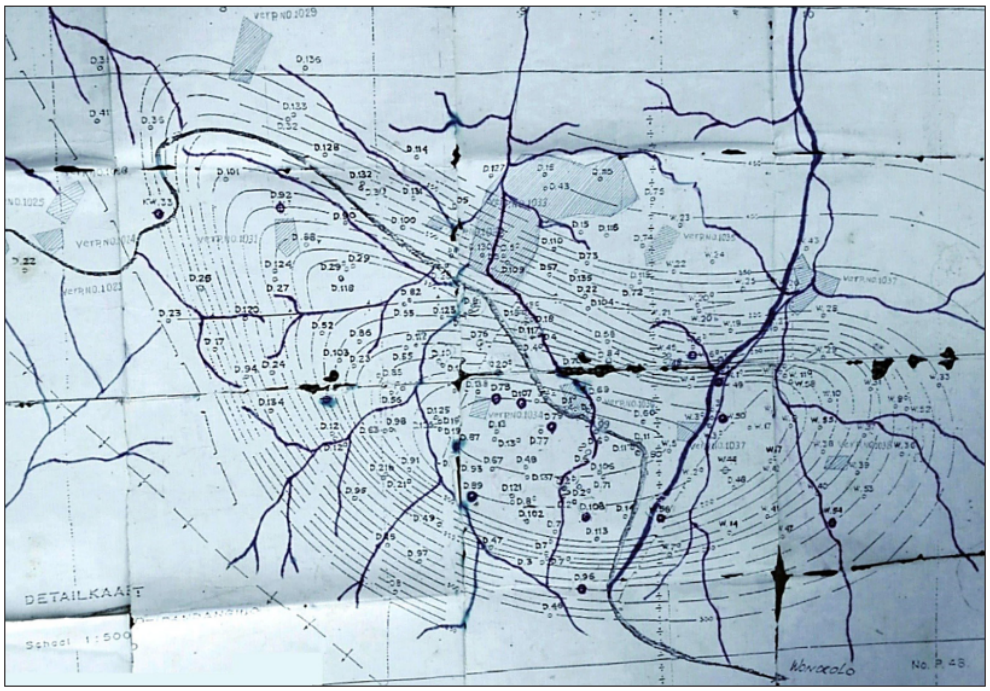

Figure 1. Crude oil drilling points during Dutch Governance period at Wonoloco sub-district

The depth of crude oil wells ranged between 200–400 m but they could be utilized only for 50–150 m, because of the limited capabilities of the truck and slick line engines which are only able to pull a thimble full with crude oil at these depths [Marwoto 2010]. The thimble is a steel-shaped pipe with a valve at the end. When the thimble is lifted to the surface, the valve would open and crude oil would spray out. Then, crude oil would be going down into a pond and separated with carried water using a small container manually. Most of the crude oil is collected to the collecting station and would be shipped to Pertamina for processing into fuel. In addition, some of the crude oil is simply refined by heating on an underground fireplace for 6 hours to form half-boiled oil. The people call it 'heavy solar', and it could be used as fuel for truck's engine, fishing boat, or agricultural equipment [Marwoto 2010, Naumi and Trilaksana 2015]. The process of crude oil exploitation in Wonocolo public mining is presented in Figure 2.

Utilization of simple equipment with insufficient procedures during crude oil exploitation activities increases the spills potential [William et al. 2005, Alinnor and Nwachukwu 2013, Pinedo et al. 2013], which could contaminate the soil and surface water at mining area (Fig. 2). The contamination caused ecological damage of soil and surface water, because it contains petroleum hydrocarbons, measured as total petroleum hydrocarbons (TPH), which were toxic, mutagenic, and carcinogenic [Liu et al. 2012, Wang 2011]. TPH consists of aliphatic and aromatic hydrocarbons that are composed of C_{5-35} chains with various structural carbon atom configurations. The more complex the atomic structure is, the more hydrophobic the hydrocarbon compounds would be [William et al. 2005]. It made the TPH became hardly degradable for microorganisms and persistent in soil and water [Nicolotti and Egli 1998, Chorom and Hosseini 2011., Das and Chandran 2011].

On the basis of the impact to environment, US-EPA [U.S. Environmental Protection Agency 2009] classified TPH as a priority pollutant. Several states and countries such as Texas, Lousiana, Colorado, Michigan, and Indonesia determine the maximum limit of TPH in soil as 10,000.00 µg/g [Agency for Toxic Substances and Disease Registry 1999, Hamilton and Sewell 1999, Indonesian Environment

Figure 2. Crude oil exploitation facility in Wonocolo public oil fields in April 2017

Ministry Decree 2003] but there is no specific standard for the surface water [Akropido and Onianwa 2015]. An earlier study [Handrianto et al. 2012] has reported that the soil at Wonocolo public oil fields has been polluted because it contains high concentration of TPH reaching 41,200.00 µg/g. Although the soil has been polluted, the investigation of TPH distribution and pollution level in soil and surface water has been unknown until now. The aims of this research were to determine the pollution level and migration of TPH in soil and surface water at Wonocolo public crude oil fields. It is necessary to determine the required cleaning-up levels and to suggest remediation alternatives to minimize the risk [Riccardi et al. 2013].

MATERIALS AND METHODS

Soil and surface water sample collection

The soil samples were collected in April 2017 from three contaminated sites, according to the crude oil exploitation activities. Those were old wells (OW), transportation line (T), and refinery (R) areas. A soil sample was also collected from the uncontaminated site (C). The location of the soil sampling is presented in Figure 3. 114 soil samples were taken from the depths of 0–30 cm, 30–60 cm, and 60–90 cm at all sampling location. Rocks and other particles were removed from soil samples and mixed to make twelve composite samples, then transferred to HDPE containers [Margesin and Schinner 2005].

Meanwhile, two surface water samples were collected from a nearby river intersection at Wonocolo public oil fields area (see Fig. 3). According to Indonesian National Standard No. 6989.57 [Indonesian National Standard 2008], the water samples were taken from the middle part of river depth, at different distances from the river side using sterile glass bottles. The samples were mixed to obtain a composite sample, and placed in sterile glass bottle. All samples were stored at 4°C in the dark and transported to the laboratory [Margesin and Schinner 2005, Indonesian National Standard 2008].

Soil physicochemical analysis

The physicochemical characteristics of soil were required because it was affected by the presence of TPH [Abdel-Moghny et al. 2012]. Texture, porosity, and permeability coefficient of soil were determined by ASTM D422–90; D2216–80 and D854–00, respectively [Indonesian National Standard 1990, 2002, 2008]. The carbon and nitrogen (C/N) ratio was measured by calculation of total nitrogen [Walkley and Black 1934] with organic carbon content [ASTM 2011].

TPH Analysis

Prior to conducting analysis, the soil samples were homogenized and sieved through a 2-mm mesh sieve, and then stored in a refrigerator at 4°C. Twelve grams of homogenous soil samples were extracted in duplicate using soxhlet method with 120 mL hexane (Merck, Germany), following the standard of APHA-AWWA and WEF No. 5520D [APHA-AWWA-WEF 2005]. Extraction was also performed for surface water sample (500 mL) by adding 100 mL of hexane separated by a separating funnel, in accordance with the standard No. 5520B by APHA AWWA and WEF APHA-AWWA-WEF 2005]. In order to obtain the hydrocarbon extract, silica gel (Merck, Germany) was added into crude oil extracts to remove water and polar

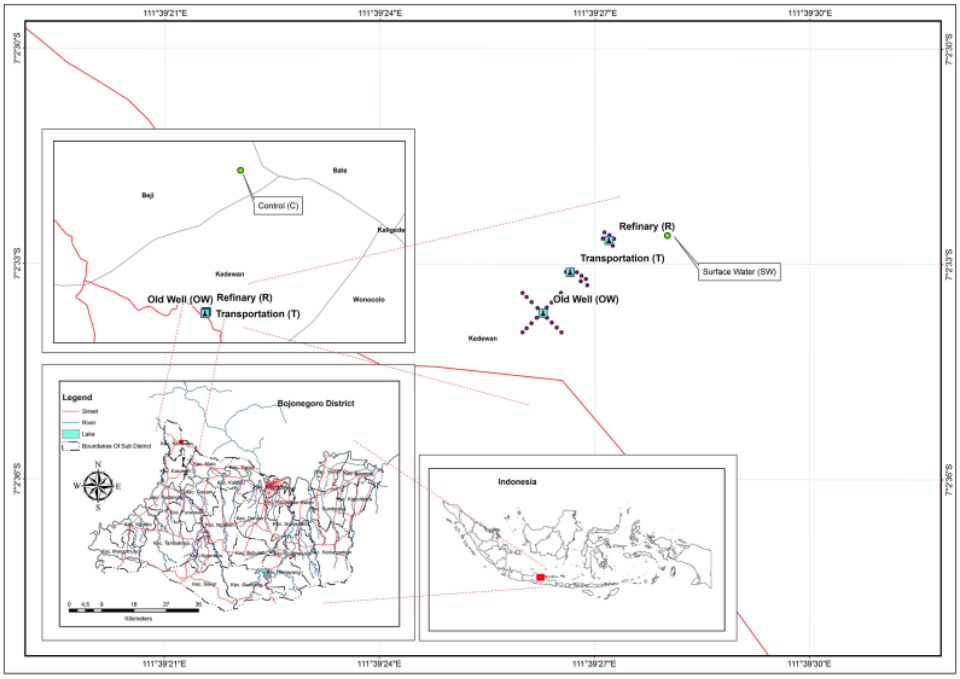

Figure 3. Soils and surface water sampling location in Wonocolo public crude oil fields

compounds. The crude oil extracts were stirred for 5 minutes using magnetic stirrer and separated by filter paper (Whattman No. 42). Then, the hydrocarbon extracts were evaporated using water bath and concentrated to 1 mL according to APHA-AWWA and WEF standard No. 5520F [APHA-AWWA-WEF 2005].

The hydrocarbon extract was used for TPH analysis which was performed using FT-IR spectrometer (Thermo Scientific™ Nicolet iS5). The absorbance was recorded at 2930 cm^{-1}, according to ASTM D7066–4 [ASTM 2011] and reached the hydrocarbon peaks. The measurement was performed by internal calibration method and TPH analysis was observed by comparison of their peaks with standards. All samples were analyzed in triplicate to ensure precision and accuracy of the analytical method with error rate and recovery efficiency between 5–15% and 80–95%, respectively. The procedural blank was determined by going through the extraction and clean-up procedures using glass beads instead of a soil sample. The results indicated that they were generally low and posed no problem to the analytical quantification.

RESULTS AND DISCUSSION

Physicochemical characteristics of soil

The characteristics of soil from four sampling locations at all depths are listed in Table 1. The results showed that soil pH was in neutral range of 6.60–7.20. It is similar to soil pH in one of China's oil fields of 6.80–8.20 that were reported by Wang et al. [2011]. These values indicated that crude oil spill can affect the soil pH balance. Cation exchange [Ying et al. 2013, Sari et al. 2016], as described by Osuji et al. [2006] resulted in that the cations in soil such as Ca^{2+} and Mg^{2+} have been able to exchange with anions and then created a pH balance.

Crude oil spills also affect the C/N ratio of soil. As shown in Table 1, various values were obtained in the analysis of soil C/N ratio; the highest was found in OW (35.07) and R (25.85) at 0–30 cm depth. Then, C/N ratio value at OW and R decreased initially as the depth increased (see Table 1). The high C/N ratio was caused by crude oil accumulation at the upper layer due to continuous spillage [Iwegbue et al. 2003, Abioye 2011, Kurnaz and Büyükgüngör 2016]. Further-

Table 1. Physicochemical characteristics of Wonocolo public crude oil fields soil

Soil(s) source	Parameters					
	pH	C/N ratio	Soil texture	Permeability (cm/min)	Porosity (%)	Saturation degree (%)
OW						
0–30 cm	6.60	35.07	Silty clay	5.12E-10	53.09	67.74
30–60 cm	7.20	31.86		2.74E-11	55.33	64.53
60–90 cm	6.90	23.54		1.29E-10	56.36	68.62
T						
0–30 cm	7.00	9.72	Silty clay	3.44E-08	57.50	62.77
0–30 cm	7.00	10.42		7.26E-08	58.07	61.23
30–60 cm	7.00	10.30		1.39E-09	56.92	67.69
R						
0–30 cm	6.90	25.85	Silty clay	4.79E-10	52.74	73.49
30–60 cm	7.00	16.61		5.69E-10	52.75	69.91
60–90 cm	7.20	14.28		9.09E-11	52.65	71.14
C						
0–30 cm	7.00	11.49	Silty clay	3.47E-10	55.22	53.18
30–60 cm	7.00	13.47		9.00E-08	58.88	53.5
60–90 cm	7.00	11.38		2.39E-08	55.58	70.25

more, Abioye et al. [2011] described that crude oil has long carbon chains, which would affect the carbon content in the soil. In contrast, T has lower C/N ratio because slightly oil spills occurred while C was an uncontaminated soil.

Crude oil accumulation in the upper layer of soil is influenced by the soil texture. According to the USDA standard, the soils were classified as silty clay with low permeability ($2.39 \cdot 10^{-8}$ to $9.09 \cdot 10^{-11}$ cm/min) although the porosity was high between 52.65–58.88%. High porosity might reduce the soil permeability due to the pores clogging by crude oil [Abosede 2013]. This is explained by the fact that the physical characteristics of soil could affect the contamination of crude oil.

Concentration and migration of TPH in soils

TPH concentration in twelve composite soil samples collected from Wonocolo public oil fields were varied, as listed in Table 2. TPH concentration in OW soil at the depths of 0–30 cm, 30–60 cm, and 60–90 cm were the highest, i.e 107,189.63; 7,006.71; and 4,529.14 µg/g, respectively. High concentration of TPH also found in R soil that were 76,752.46; 839.61; and 559,87 µg/g for the depths of 0–30 cm, 30–60 cm, and 60–90 cm, respectively. Similarly to C/N ratio value, the high concentration at both soils was also caused by numerous oil spills that could be seen through the degree of saturation. In accordance with Hardiyatmo [2012], the values of saturation degree that ranged from 64.53 to 73.49% were classified as high humidity. High concentration of TPH is also due to petroleum hydrocarbon that could be associated with sorbed organic matter in the soil [Abdel-Moghny et al. 2012]. At the same depths, the following TPH concentrations in T soil were obtained: 52,328.14; 2,256.30; and 1,919.19 µg/g. In contrast, the lowest concentrations were observed in soil C, in total of 159.94; 119.80; and 139.87 µg/g, respectively.

The results showed that the top soil (0–30 cm) contained high concentration of TPH (52,328.14 to 107,189.63 µg/g). Compared to the maximum limit of TPH in soil of 10.000 µg/g, it could be concluded that the topsoil at three soil sampling locations were polluted. Lower limit concentration of TPH presented by environmental guidelines and standards for the petroleum industry in Nigeria was 5,000 µg/g [DPR 2002]. Therefore, the pollution was found at OW (0–30 and 30–60 cm), T (0–30 cm), and R (0–30 cm) soils. If the Mexican legislation limit of TPH was 2,000 µg/g [Iturbe et al., 2008] is taken into account, then polluted soil was found at OW (all depths), T (0–30 cm and 30–60 cm), and R (0–30 cm). On the other hand, the Italian regulation set a lowest maximum limit of TPH by 50 µg/g [Riccardi et al., 2013], which indicated OW, T, R, and C areas from the surface up to 90 cm below are completely polluted. Soil C containing TPH might

Table 2. TPH concentration in crude oil contaminated soils

Soil depth (cm)	Concentration (µg/g)									
	Wonocolo public crude oil fields				Crude oil fields on river states of Nigeria	Shell marginal crude oil pipeline at south Niger Delta, Nigeria		Crude oil exploitation site at Yellow River Delta, Shandong province, China	Crude oil spill site in Liaohe oil field, Liaoning province, China	Crude oil spill site in Batman, Southeast Anatolia of Turkey
	OW	T	R	C						
0–15	107,189.6	52,328.1	76,752.5	159.94	73.17–1534.66	11±6 to 402±3	9 to 289±3	-	-	-
0–30					-	9±13 to 337±5	11 to 413±7	-	11,400-21,800	-
0–50	-	-	-	-	80.15–1438.00	-	-	-	-	-
30–60	7,006.71	2,256.30	839.61	119.80	-	13±7 to 184±9	13 to 178±11	-	-	-
60–90	4,529.14	1,919.19	559.87	139.87	-	-	-	-	-	-
50–100	-	-	-	-	-	-	-	-	-	-
No specific depth	-	-	-	-	-	-	-	18,000.0	-	48,300.0
Reference					Alinnor and Nwachukwu 2013	Okop et al. 2013	Okop et al. 2012	Wang et al. 2011a	Xu and Liu 2010	Kurnaz and Büyükgüngör 2016

be caused by hydrocarbon deposition from the air, due to its semi-volatile characteristics.

As shown in Table 2, TPH concentration in soils has decreased as the depth increased that might be affected by low soil permeability [Iwegbue et al. 2003]. Low permeability of soil was an advantage, as crude oil has low potential migration and risk of groundwater contaminated [O'Reilly et al. 2001]. The statement was supported by oil vertical transport prediction in OW area using Raoult's Law [Trihadiningrum 2016] that crude oil takes up to 2.8 million years to reach the depth of 2 m below the topsoil. These calculations used the value of permeability, porosity, density from OW properties (see Table 1), and distribution coefficient of crude oil was 0.90 m^3/kg [Rios et al. 2015].

Table 2 shows that petroleum hydrocarbon concentration from crude oil which contaminated soil in present work is significantly higher than these of other studies. For example, in Nigeria it was found several sites that have been contaminated by TPH to 1,534.66 µg/g [Okop et al. 2012, 2013]. Higher concentrations have been found in some areas in China and Turkey, ranging from 11,400 up to 21,800 µg/g [Xu and Liu 2010.] and 48,300 µg/g [Kurnaz and Büyükgüngör 2016], respectively. Contamination of TPH in soil that has been reported was caused by crude oil spills and leaks which continuously occurred for long periods of time. The abundance of crude oil in the soil made the TPH hardly degraded through the metabolic process of microorganisms because the half-life in the soil ranges between 12–298 days [Park and Park 2010].

Soil pollution levels in Wonocolo public crude oil fields

In Indonesia [Indonesian Government Regulation 2014], TPH pollution levels are divided into several categories, i.e. A (40,000 µg/g), B (5000 µg/g), C (1000 µg/g). US-EPA [U.S. Environmental Protection Agency 2009] also classifies the TPH pollution levels into three categories: A, B, and C with different concentration of ≥ 40,000; 10,000–40,000; and 1,000–10,000 µg/g, respectively. Meanwhile, pollution category in New Jersey and several European countries like Netherland and Spain are seriously contaminated soil by 5,000 µg/g TPH [VROM 2012, NJDEP 2008]. The comparison of soil pollution level in Wonocolo crude oil public mining with several categories is listed in Table 3.

Pollution of TPH at Wonocolo public crude oil fields reaches the highest level at 0–30 cm depth (see Table 3), which were classified as seriously contaminated and the highest hazard for humans (category A). According to [Indonesian Government Regulation 2014, VROM 2012], category A means the remediation is required to reduce the hazard level of TPH until the concentration is permitted to be disposed at a landfill or other licensed facility (catego-

Table 3. TPH soil pollution level

Soil(s) source	TPH pollution level							
	Indonesian Government regulation No. 101 [2014].			US-EPA [2009]			Netherland [VROM 2012]	New Jersey [NJDEP 2008]
	A	B	C	A	B	C	Serious	
	40,000 µg/g	4,000 µg/g	1,000 µg/g	<40,000 µg/g	10,000–40,000 µg/g	1,000–10,000 µg/g	5000 µg/g	
OW								
0–30 cm	√	-	-	√	-	-	√	
30–60 cm	-	√	-	-	-	√	√	
60–90 cm	-	√	-	-	-	√	-	
T								
0–30 cm	√	-	-	√	-	-	√	
30–60 cm	-	-	√	-	-	√	-	
60–90 cm	-	-	√	-	-	√	-	
R								
0–30 cm	√	-	-	√	-	-	√	
30–60 cm	-	-	-	-	-	-	-	
60–90 cm	-	-	-	-	-	-	-	
C								
0–30 cm	-	-	-	-	-	-	-	
30–60 cm	-	-	-	-	-	-	-	
60–90 cm	-	-	-	-	-	-	-	

ry B and C). Thus, the remediation process at Wonocolo public crude oilfields could be performed on topsoil to prevent further crude oil percolation into soil and also to avoid TPH and soil matrix interaction that can cause higher pollution loads. Indonesian Environment Ministry Decree No. 128 [Indonesian Environment Ministry Decree 2003] suggest that remediation for TPH polluted soil less than 150,000 µg/g could be conducted through a biological process such as composting.

TPH-polluted soil potential risk to human health

The information on toxicity of TPH-polluted soil is important for the human health exposure in Wonocolo public crude oil fields, but it has been unknown until now. According to Park and Park [2010], the TPH potential exposure pathways to human are through inhalation and dermal contact with soil. Furthermore, the DPR [Department of Petroleum Resources 2002] stated that TPH toxicity in soil has established the concentration exceeding 1,000 µg/g. The standard could be used as a reference to evaluate the human health risk for TPH polluted soil in Wonocolo public crude oil mining. Pinedo et al. [2013] described that there was a correlation between TPH concentration and potential human health risks. As listed in Table 2, TPH concentration in OW (all depths), T (all depths), and R soils (0–30 cm) was found to be higher from the standard indicating that it carried human health risk, such as the carcinogenic potential [Park and Park 2010].

Concentration of TPH in surface water

TPH concentration from a surface water sample that was collected from a small river in Wonocolo public crude oil mining was 211,025.73 µg/L. The result suggested that TPH contamination is caused by crude oil spills and residue disposal to the river from drilling and refinery activities, respectively. Crude oil was released to water and formed a layer at the surface. This situation would inhibit the diffusion of oxygen (presented in Figure 4). The limitation of oxygen in water cause anoxic-anaerobic condition to form. Under the anoxic-anaerobic conditions, TPH degradation by microorganism metabolic process became limited. In addition, the half-life of TPH in water ranging from 4 up to 226 days, caused an accumulation [Park and Park 2010].

Figure 4. Surface water condition at Wonocolo public crude oil fields on April 2017

TPH concentration in the surface water was higher than in several studies at other locations. Olufemi et al. [2011] reported that TPH concentration at Ubeji River, Warri, Nigeria was 73,500.00 µg/L. Daniel and Nna [2016] also reported that Cross River Estuary, Niger Delta has slightly lower concentration of TPH ranging between 13,161.81 to 24,854.62 µg/L. A similar concentration of TPH was found by Clinton et al. [2009] in oil-polluted mangrove wetland in the Niger Delta of 23,600.00 µg/L. Lower concentration of TPH was reported by Akporido and Onianwa [2015] in Esi River, Western Niger Delta that ranged from 2,510.00 up to 5,630.00 µg/L.

Until now, TPH limit standard regulation for surface water could not be found in Indonesia or other countries. The regulation is only available for selected waters. DPR [Department of Petroleum Resources 2002] recommendation for TPH concentration in groundwater was 600.00 µg/L. Furthermore, Van der Leedeen et al. [1990] reported that TPH limit concentration for recreational water was 5,000.00 µg/L. All standards were below the TPH concentration in surface water at Wonocolo crude oil public mining, indicating that it is polluted. Thus, the water was not recommended for use.

CONCLUSION

The public crude oil mining activities in Wonocolo sub-district have caused TPH pollution in soil and surface water. The highest TPH concentration in OW, T, and R soil samples were observed at the depth of 0–30 cm (52,328.14 – 107,189.63 µg/g) because they had very low permeability of $3.44 \cdot 10^{-8}$ to $2.74 \cdot 10^{-11}$ cm/s. This result confirmed that these sites were polluted by TPH and classified as highest hazard level (category A) which means the top soils needed a remediation. The remediation can be conducted using a biological method. Meanwhile, the concentration of TPH in surface water was high, amounting to 211,025.73 µg/L. There was no standard for TPH concentration in surface water, thus the level of contamination was unknown.

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PAGE 1

PAGE 2

PAGE 3

PAGE 4

PAGE 5

PAGE 6

PAGE 7

PAGE 8

PAGE 9

PAGE 10

PAGE 11