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RESEARCH PAPER

Application of nanotechnology for remediation drill cutting soil

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A B S T R A C T:

This study was carried out for remediation of drill cutting soil from two oil field sites in Kurdistan Region of Iraq. nanotechnology was implemented. Heavy metal (Pb, Cd, Cr, Ni and petroleum hydrocarbons (C8-C24, C24-C40, TPH) were analyzed before and after remediation drill cutting soil. The experiment was designed. The results indicate that the concentration of heavy metals (Ni, Cd and Cr) was within the normal range except the concentration of Pb. The result showed that the removal rate of heavy metal by nano technic was (Pb%16, Ni %57, Cd% 48, Cr%38). And the removal rate of hydrocarbons was (TPH%57, C8-C24%62, C24-C40%41). these means the (nZVI) has effect for minimizing heavy metal and petroleum hydrocarbons in drill cutting soil.

KEY WORDS: drill cutting, heavy metal, nanotechnology, petroleum hydrocarbons
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1.INTRODUCTION :

Unfortunately, oil and gas are one of the world's most important resources and play a major part in the world economy, but all operations associated with petroleum exploration and production have an impact on the environment in some way (Lodungi et al., 2016). Toxic compounds in oil and gas well drilling waste discharges are potentially damaging to the ecology. Drilling waste is one of the most significant amounts of waste produced during oil and gas exploration and production.(Ismail et al., 2017) The drilling operation generates two significant wastes: drilling fluid waste and drilling cuttings. The volume of drilling waste generated by each well varies depending on the depth and diameter of the wellbore (Onwukwe et al., 2012). Drilling Fluid (Drilling Mud): This fluid is utilized during the drilling of oil wells and is generally consisting of natural and synthetic different chemicals.

Drilling fluids provide a variety of functions, including lubricating and cooling the drill bit and drilling equipment, suspending and releasing drill cuttings to the surface, sealing permeable formations, and decreasing corrosion rates (Dhiman, 2012). Drilling muds are classified into three types: water-based drilling muds (WBDM), oil-based drilling muds (OBDM), and gas-based drilling muds (Mikos-Szymańska et al., 2018). The present study focuses on WBDM which depends on water as a basic material in its composition and contains other chemical additives. This type of drilling mud is typically a favorable mud for environment because it has not major toxic compounds, however contains some of hydrocarbons, heavy metals and chemical additives. The concentration of hydrocarbons and these metals depends on the type of drilling mud. Petroleum hydrocarbons and Heavy metal concentrations in cuttings vary depending on the kind of drilling fluid used, the additives utilized, and the naturally geological of the formation being drilled. They are rock pieces generated by cutting rock layers and transported to the surface by drilling mud(Amin et al., 2018, Mikos-Szymańska et al., 2018).hydrocarbons and these products are used in petroleum exploration

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which are found in and have been designated as a main environmental contaminant by the US Envir onmental Protection Agency, are mutagenic and c arcinogenic. PAHs are the most tenacious and hazardous type of soil contaminants found in crude oil. After entering the soil, PAH get trapped in the soil pores and are held by the soil matrix. As a result, removing them from the soil is extremely difficult. Because of these alterations and potential threat to public health and the environment(Ahmad et al., 2020).Heavy metals are some of the major contaminants in impacted spent drilling mud, that cause harm and pollution to the environment. The mixing of wastes containing high concentrations of heavy metals with soil can transfer large amounts of these minerals to the plant, threatening human life and the other organisms that feed on these plants(Felix et al., 2018).When the harmful are released into the environment, they can be leached into the underground waters, depositing in the aquifers, or run off into surface waters and soils thereby resulting in water and soil pollution. Thus, petroleum hydrocarbons and heavy metals become a potential contaminant for environment that can partake in trophic transfer in food chains (Morais et al., 2012, Singh et al., 2011, Ahmad et al., 2020). Remediation method that used in present study for minimize environmental and human health impact was Nano remediation by zero valence iron nanoparticle (nZVI). Iron-based remediation techniques which include ecological remediation are highly being evolved and examined at various scales in the lab and in the site(O'Carroll et al., 2013). Zero-valent iron nanoparticles are strong reducing agent and very good adsorbent for various pollutants (Cundy et al. 2008). They are widely used to remove heavy metal such as Mercury ($Hg2^{+)}$, Nickel ($Ni2^{+}$), Cadmium ($Cd2^{+}$), Lead ($Pb2^{+}$) and Chromium ($Cr6^{+}$) from contaminated soil. The major benefit of iron is that is the widely available metal on the planet, making sorbents made of it really cost efficient. Furthermore, iron metal is non-toxic in its elemental nature, making it environmentally safe (Noubactep et al., 2012). Many oils and gas production, agricultural, and other industries have grown in Iraq's Kurdistan Region during the previous decade, some of these industries have potentially released enormous amounts of contaminants into the environment, including heavy metals. Thus, the main aim of this study is minimized potential impact to environment and human from drill cutting soil. For minimizing these impacts from drill cutting on the human and environment have some techniques for remediation cutting such as nanotechnology.

2.MATERIALS AND METHODS

2.1. Sample collection and location

Drill cutting sample were collected at depth of 0-60 cm randomly in September 2021 from two oil fields in the kurdistan region - Iraq. the drill-cuttings put into a clean and labeled polyethylene bag and stored in refrigerator for further laboratory analyses(Ahmad and Ganjo, 2020).

2.2 Prepared sample

The soil samples were air dried for 72 hours, then crushed by wood rod and sieved through 2 mm sieve for removed large particle. nano remediation was implemented in this study as follow:

2.2.1 Nano remediation

100g of sixteen subsample from Soil cutting sample were taken , then mixed with (0.2,0.4,0.6 and 0.8 gm) zero valent iron nonpowdered APS:25nm with and then added 7ml distilled water in to mixture moisture content of the soil mixture was contain 7.06% then incubated at 25ºC for a week, as described by (Sun et al., 2020, Zhang et al., 2010, Kumpiene et al., 2006), Murgueitio et al. (2018), (Felix et al., 2018). The experimental design based on Factorial CRD.

2.3 Sample analysed

The heavy metal in soil cutting was extracted by using 0.05M EDTA method as described by (Sabienë et al., 2004), then heavy metal determined by A.AS (Atomic absorption spectrophotometer),However the Gas chromatography (EPA method number 8015) and an HP 6890 GC-MS ,GC part: Agilent 7890,Mass part: Agilent 5975,Column:Bp-5Capillary Column, were used to measure the TPH concentration.

2.4 Contaminant removal indices

Percentage removal rate of contaminant used to distinguish the volume of the removed contaminant between heavy metals and petroleum hydrocarbons in drill cutting soil (Francy et al., 2020).

R = (Ci−Cf) / Ci × 100

Where R is the removal rate of contaminant (percentage), Ci is the initial contaminant concentration, and Cf is the final contaminant concentration.

2.5 Statistical analyses

The experimental design base on a factorial completely randomized design (Factorial CRD). Data were statistically analyzed by using SPSS version 25. the difference between the treatments means were compared by applying Duncan multiple comparison tests at the level of significant (5%)(Townend, 2013).

3. RESULTS AND DISCUSSION

3.1. Mean of some heavy metals in cutting soil from two different sites

The chemical characteristics of drilling waste depend largely on geological factors, and on different drilling techniques used at the well sites, especially the type of muds. Data present in table (1) showed that (nZVI) levels significantly at (0.05) effect on the heavy metal in cutting soil from two sites. The maximum values were (104.83, 0.297, 1.97) (0.038) mg/kg for (Pb, Ni, Cr) were recorded from (S1) except (Cd) which was recorded from (S2) respectively. While the minimum values were (94.59, 0.115, 1.368) (0.030) mg/kg for (pb, Ni, Cr) were recorded from (S2) except (Cd) was recorded from (S1) respectively. The results indicated that the composition of these heavy metals have different between (S1) and (S2). The reason may be due to the different geological formation, geographical factor and physiochemical properties of cutting soil, different drilling technique, different chemical additive that used for drilling mud of these sites area. These results and interpreting agreed with those have been reported by (Jamrozik et al., 2019).

Table (1) Mean of some heavy metals in cutting soil from two different sites

Site	Pb	Ni	Cr	Cd
Site 1	104.83	0.297	1.972	0.030
Site 2	94.59	0.115	1.368	0.038
LSD 0.05	3.989	0.111	0.523	0.000

3.1.1Effect Nano remediation on the drill cutting soil

3.1.1.2Effect of (nZVI) on the concentration heavy metal in drill cutting soil

The data analysis in table (2) indicated that concentration of (nZVI) significantly at (0.05) affected on some heavy metals in drill cutting soil. The highest value was (149.63, 0.4393, and 2.54695) (0.06291) mg/kg for concentration heavy metal in drill cutting soil for (Pb, Ni and Cr) recorded from the control (C1) except the concentration (Cd) recorded from treatment (C2), respectively. While the lowest value (125.99), (0.2012) and (0.0253 ,1.7170) mg/kg for same heavy metals (Pb), (Ni) and (Cd, Cr) were recorded from treatment (C4), (C2), (C5, C5), respectively. The result indicates that the increasing concentration of (nZVI) does not effect of decreasing of concentration heavy metal in soil cutting. whereas, most of the heavy metals concentration decreased by using (nZVI) except (Cd) increasing concentration by increasing (nZVI) concentration from treatment (C2). iron based technologies for remediation of contaminated soil has been documented (Henn et al., 2006). nZVI has been proven to be a strong chemical reductant and is able to convert many mobile into immobile forms(Cundy et al., 2008) .These result and interpretation are similar to that recorded by (Felix et al., 2018) reported that (nZVI) can effectively treated drilling mud with heavy metal. These results were confirmed by the recording significant correlation between concentration of (nZVI)and concentration heavy metals in drill cutting soil, The correlation coefficient values attained (r: - 0.9037, -0.7326) for Pb and Cd respectively. Figure (1&2) supports negative correlation between the concentration of (nZVI) and concentration of (Pb, Cd). Highest value for efficiency removal rate of concentration heavy metal by using(nZVI) (%16, %57, %38, %48) for (Pb, Ni, Cd, Cr) from treatment (C4, C2, C3, C5), respectively. Figure (3) supports relationship between concentration of (nZVI)and removal rate of heavy metal in cutting soil from.

Table (2) Effect of (nZVI) on the concentration heavy metal in drill cutting soil

Concentration	pb	Ni	Cd	Cr
C1	149.63[a]	0.4393[a]	0.0599[a]	2.5469[a]
C2	128.52[b]	0.1906[a]	0.06291[a]	2.2250[a]
C3	130.39[b]	0.2012[a]	0.0372[ab]	2.0136[a]
C4	125.99[b]	0.2367[a]	0.0416[ab]	2.6041[a]
C5	128.57[b]	0.3010[a]	0.0253[b]	1.7170[a]

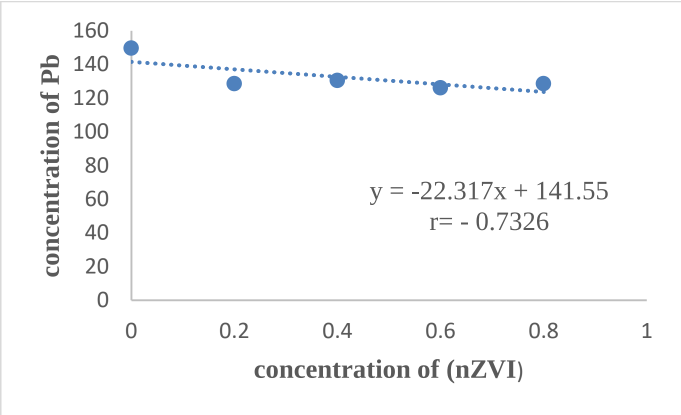

Figure (1) relationship between concentration of Pb and concentration of (nZVI)

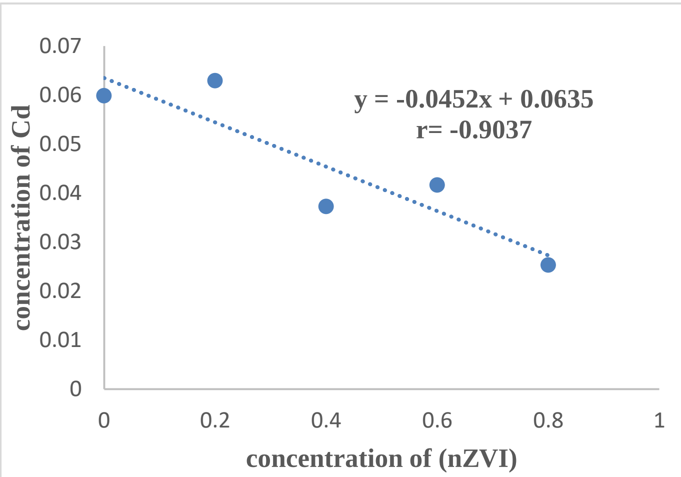

Figure (2) relationship between concentration of Cd and concentration of (nZVI)

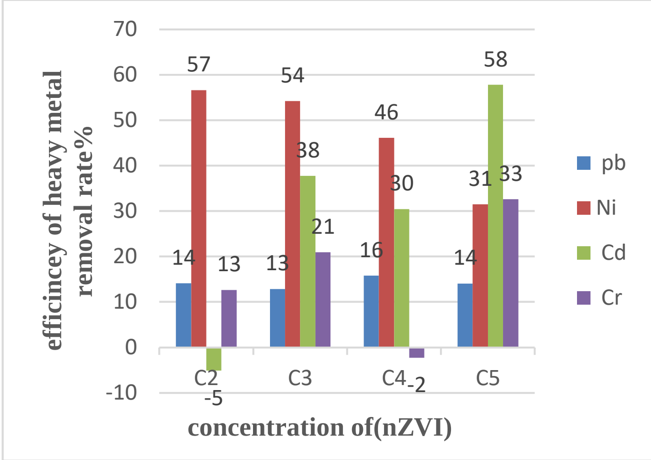

Figure (3) relationship between concentration of (nZVI)and removal rate of concentration heavy metal in cutting soil.

3.1.1.3 Effect of (nZVI) on the site concentration heavy metal in drill cutting soil

Table (3) indicated that the concentration of (nZVI) significantly level at (0.05) affected on the concentration of site heavy metals. The result showed that the highest values were (168.44, 0.77, 2.95) (0.8) mg/kg of heavy metals (Pb, Ni, Cr), (Cd) were recorded from (S1C1) except (Cd) recorded from (S2C1), respectively. While the lowest values were (121.25, 0.10, 0.02, 1.41) mg/kg of heavy metals (pb, Ni, Cd, Cr) were recorded from treatment (S2C4, S2C1, S2C5 and S2C3), respectively. Increasing concentration of (nZVI) does not effects of decreasing heavy metal in soil cutting at both sites. however, most of the heavy metals concentration decreased by using (nZVI) whereas concentration (Ni)increased by increasing of concentration of (nZVI) in cutting soil from both sites. This result indicates that the concentration of (nZVI) have effect on the concentration of heavy metal from both sites, thus may be due to, thus may be due to Elemental iron slowly oxidizes to ferrous iron and releases two electrons. These electrons begin to function in a variety of reactions that lead to the transformation of target contaminants such as heavy metal (Tratnyek and Johnson, 2006).

Table (3) Effect of (nZVI) on the site concentration heavy metal in drill cutting soil

Site concentration	heavy metal concentration in Cutting soil mg/kg			
	Pb	Ni	Cd	Cr
S1C1	168.44[a]	0.77[a]	0.04[abc]	2.95[a]
S1C2	132.52[b]	0.25[b]	0.05[abc]	2.92[a]
S1C3	133.48[b]	0.27[b]	0.03[c]	2.62[a]
S1C4	130.82[b]	0.31[b]	0.04[abc]	2.69[a]
S1C5	131.89[b]	0.38[b]	0.03[bc]	1.94[a]
S2C1	130.81[b]	0.10[b]	0.08[a]	2.15[a]
S2C2	124.51[b]	0.13[b]	0.07[ab]	1.53[a]
S2C3	127.31[b]	0.14[b]	0.04[abc]	1.41[a]
S2C4	121.16[b]	0.17[b]	0.04[abc]	2.52[a]
S2C5	125.25[b]	0.22[b]	0.02[c]	1.50[a]

3.2 Mean of petroleum hydrocarbons in drill cutting soil from two different sites

Data present in table (4) indicated that (nZVI) significantly at (0.01) effected the petroleum hydrocarbons in drill cutting soil from two sites. The highest values were (103.64), (364.29, 467) ppm for (C24-C40) (C8-C24, TPH) were recorded from (S1 and S2), respectively. This result indicate that have different significant between (S1 and S2), this may be due to geological formation, geographical factor, or may be due to physiochemical properties of cutting soils and on different drilling techniques used at the well sites or different using chemical additive for the type of muds. These results and interpreting agreed with those have been reported by(Jamrozik et al., 2019)

Table (4) Mean of petroleum hydrocarbons in drill cutting soil from two different sites

Sites	C8-C24	C24-C40	TPH
Site1	292.03	103.64	395.71
Site 2	364.29	102.71	467
LSD 0.01	10.841	6.051	7.095

3.2.1 Effect of (nZVI) on the petroleum hydrocarbons

3.2.1.1 Effect of the concentration (nZVI) on the concentration of petroleum hydrocarbons in cutting soil

Table (5) indicated that the concentration (nZVI) at level of significantly (0.01) on the concentration of petroleum hydrocarbons in drill cutting soil. maximum values were (569.9, 137.975, 707.875) for (C8-C24, C24-C40, TPH) were recorded from control (C1), respectively. While minimum values were (218.65, 305.1250) (81.375) for (C8-C24, TPH) (C24-C40) were recorded from (C5) and (C4), respectively. The result indicates that by increasing concentration of (nZVI) decrease the concentration petroleum hydrocarbons in drill cutting soil, this decrease may be due to the degradation /transformation petroleum hydrocarbon compound in drill cutting soil. These result and interpretation were confirmed by present in Figures (5,6,7) which refer to the significant Positive correlation coefficient between concentration (nZVI) and concentration petroleum hydrocarbons in drill cutting soil, the correlation coefficient values attained (r: - 0.8588, -0.9604, -0.9722) for (C8-C24, C24-C40, TPH), respectively. Highest removal rate of petroleum hydrocarbon in cutting soil were (%62, %41, %57) for (C8-C24, C24-C40, TPH). Figure (4) supports the relationship between concentration of (nZVI)and removal rate of petroleum hydrocarbons in cutting soil. This result and interpretation are similar to the result reported by(Chang et al., 2007).

Table (5) Effect of the concentration (nZVI) on the concentration of petroleum hydrocarbons in cutting soil

concentration	C8-C24	C24-C40	TPH
C1	569.9[a]	137.975[a]	707.875[a]
C2	302.875[b]	124.05[ab]	427.025[b]
C3	306.3[b]	86[bc]	392.300[b]
C4	243.075[bc]	81.3750[c]	324.45[c]
C5	218.65[c]	86.475[bc]	305.125[c]

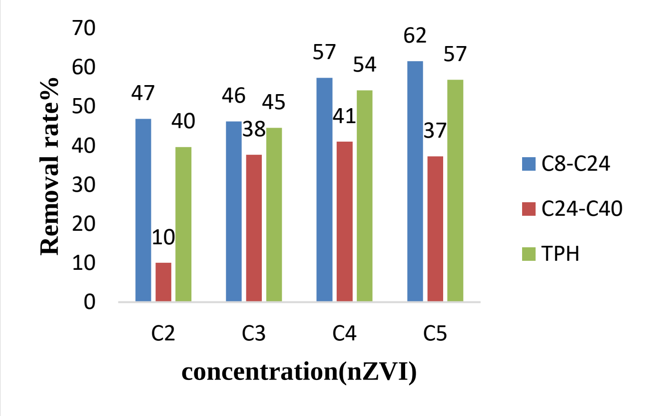

Figure (4) relationship between concentration of (nZVI)and removal rate of petroleum hydrocarbons in cutting soil

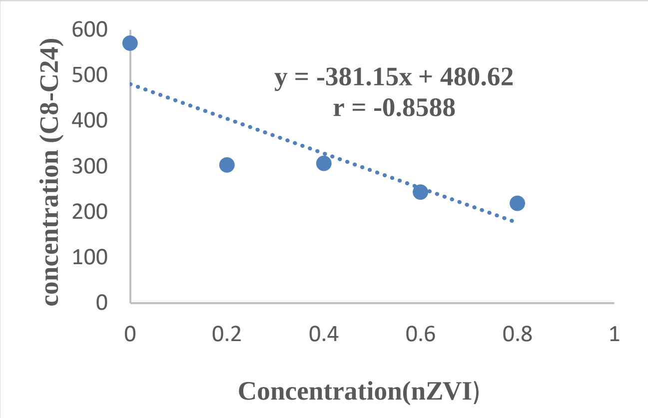

Figure (5) relationship between concentration of (nZVI) and concentration (C8-C24)

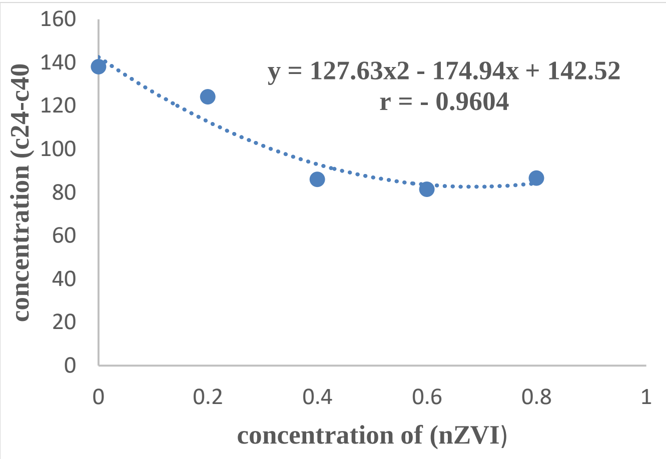

Figure (6) relationship between concentration of (nZVI) and concentration (C24-C40)

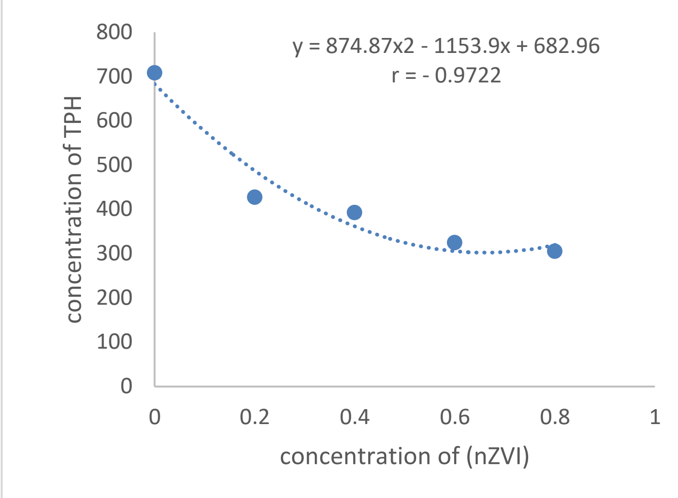

Figure (6) relationship between concentration of (nZVI) and concentration (TPH)

3.2.1.2Effect of the concentration (nZVI) on the concentration of petroleum hydrocarbons in cutting soil from two sites

Table (6) indicated that the effect (nZVI) at level of significantly (0.01) on concentration of petroleum hydrocarbons in drilling cutting soil at two sites. maximum values were (636.6,156.3, 756) for (C8-C24, TPH) (C24-C40) from (S2C1) and(S1C1), respectively. Whereas lowest values were (201.45) (70.65, 288.2) for (C8-C24) (TPH, C24-C40) from (S1C5) and (S1C4), respectively. This result indicates that the (nZVI) has effect on the decreasing of concentration petroleum hydrocarbons(ppm) in cutting soil from both sites, however petroleum hydrocarbons composition was different from both sites, this decrease may be due to the degradation /transformation petroleum hydrocarbon compound in drill cutting soil(Chang et al., 2007).

Table (6) Effect of the concentration (nZVI) on the concentration of petroleum hydrocarbons in cutting soil from two sites

Site Concentration	**C8-C24**	**C24-C40**	**TPH**
S1C1	503.2[b]	156.3[a]	660b
S1C2	299.35[d]	104.65[cd]	404cd
S1C3	238.6e[f]	98.45[cd]	337ef
S1C4	217.55[f]	70.65[d]	288.2f
S1C5	201.45[f]	88.15[cd]	289.6f
S2C1	636.6[a]	120[bc]	756[a]
S2C2	306.4[d]	143[ab]	450[c]
S2C3	374[c]	74[d]	448[c]
S2C4	268.6[de]	92[cd]	361[de]
S2C5	236[ef]	85[cd]	321[ef]

4.CONCLUSTIONS

Drilling activity in Oil exploration and production process increase some heavy metal and hydrocarbons in environment due to using some chemical during the process that have impact on the human health and environment. This study investigated remediation of drill cutting soil by using zero valance iron nanoparticle (Fe^{0}) for remediation drill cutting soil from two sites. The result showed that the zero-valance iron nanoparticle (Fe0) has effect on some heavy metals (Pb, Ni, Cd, Cr) and petroleum hydrocarbons in drill cutting soil, thus may be due to Elemental iron slowly oxidizes to ferrous iron and releases two electrons. These electrons begin to function in a variety of reactions that lead to the transformation of target contaminants such as heavy metal and petroleum hydrocarbons. removal rate of heavy metal by nano technic was (Pb%16, Ni %57, Cd% 48, Cr%38). And the removal rate of hydrocarbons was (TPH%57, C8-C24%62, C24-C40%41).

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Conflict of Interest (1)

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