Characterization, Efffects and Chemical Treatment of Heavy Metals in Produced Water from Injection Wells using Hydroxide Precipitation

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ABSTRACT

Produced water obtained from five (5) water injection Wells in the Niger Delta area Nigeria were analysed for heavy metal ion concentrations using Themo Elemental Flame Atomic Absorption Spectrophotometer (AAS). Results obtained show that concentrations of lead (Pb^{2+}), cadmium (Cd^{2+}), chromium (Cr^{2+}), nickel (Ni^{2+}), cobalt (Co^{2+}), vanadium (V^{2+}), zinc (ZN^{2+}), mercury (Hg^{+}), silver (Ag^{+}) and copper (Cu^{2}) were above acceptable limits as specified by Environmental Guidelines and Standard for Petroleum Industries in Nigeria (EGASPIN). The concentrations of As^{2+}, Fe^{2+}, K^{+} and Mn^{2+} were within specification. Commingled produced water from the five water injection Wells were subjected to hydroxide precipitation using $Ca(OH)_2$ at concentrations of 10, 20, 30, 40 and 50 ppm respectively. Results obtained show that all the heavy metal ions reduced within acceptable limit at 30 ppm chemical concentration. Heavy metal ions reduced with increase in chemical concentration with a corresponding increase in the pH of the solution which was however still within specification. The hydroxide converts the metal ions dissolved in solution into solid particles for easy sedimentation. $Ca(OH)_2$ precipitates metal ions by changing the pH and electro-oxidizing potential of the solution. Properly treated produced water can be reinjected into the reservoir to enhance oil recovery, used in agriculture for irrigation purposes, discharged into the sea and other water bodies during offshore operations or even used in drilling services. Produced water discharged into the soil are non-biodegradable so must be adequately treated before discharge. Plants pick up heavy metals through their roots by the release of a variety of root exudates which changes the rhizosphere pH of the metal ion in solution thereby making them bioavailable for plant uptake by osmosis. Heavy metals in plants can exert a variety of toxic actions by damaging plant chloroplast thereby disturbing photosynthesis. Humans take in heavy metals into their system by consuming contaminated plants, water as well as inhaling contaminated air. Consumption, ingestion and inhalation of heavy metals by humans can cause a wide range of ailments such as cardiovascular diseases, kidney related problems, neurocognitive diseases, renal damage, heart disease, coronary artery disease, lung fibrosis, nasal cancer.

Key Words: Precipitation, Filtration, Contamination, Diseases, Concentration, Environment.

INTRODUCTION

The term 'produced water' is very significant in crude oil exploration and production, it refers to salt water from underground formations obtained as a byproduct during crude oil and natural gas extraction. In older wells produced water constitute over 80% of the crude produced and their characteristics is dependent on the geological composition of the well and the age of the well[1]. Produced water are breakthrough water from impermeable rocks within the reservoir, they exist in both oil and gas reservoirs however oil reservoirs tend to produce larger quantities of water compared to gas reservoirs[2]. Produced water could either exist below the oil or gas under certain conditions within the reservoir or at the same zone with the hydrocarbons at other instances.

The management of produced water obtained from crude oil exploration is centered on the adequate knowledge of the characteristics of the produced water, this is essential in applying the adequate treatment to ensure proper usage and disposal[3]. The application and disposal of produced water depends on the quality of treatment given to it. Adequately treated produced water can be injected into the ground into an injection well between impermeable layers of rocks to prevent the pollution of surface waters[4]. The reinjected produced water helps in boosting the reservoir pressure thereby enhancing crude oil recovery. Figure 1 shows the different layers in a reservoir rock beginning with the impermeable rock seal, the gas, oil, produced water and source rock[5]. Produced water in injection wells can also be used in environmental remediation to clean up either soil or groundwater contamination, this can be achieved by inserting clean and treated water from the injection wells into an aquifer thereby changing the direction and speed of groundwater flow towards downgradient extraction wells which removes the contaminated groundwater speedily and more efficiently[6,7].

Produced water in injection wells can also be used in the cleanup of contaminated soil using an ozonation system. Complex hydrocarbons and other contaminants trapped and inaccessible in the soil can be broken down by a highly reactive gas like ozone. This process is less expensive compared to digging out the affected areas and it is useful in built-up urban environments where digging is almost impossible due to overlying buildings[8,9]. Alternative to injection wells is the direct discharge of treated produced water to receiving waters like seas, rivers, lakes etc. Produced water can also be used in agriculture for the purpose of irrigation or livestock watering in areas where water is scarce[10]. Extensive irrigation is not typical in areas where the produced water tends to be salty due to the high cost of salinity reduction and stabilization[11].

Minimally treated produced water can be used for drilling and work over operations within the petroleum industry while highly treated water can be used as industrial or even drinking water[4]. Produced water can undergo primary, secondary and tertiary treatment depending on their level of contamination, characteristics required for reinjection and other usage[1]. Primary or physical treatment of produced water are often made possible by mechanical installations in the process while secondary or biological treatment involves the elimination of organic matter in produced water by oxidation or the process of incorporating them into the cells of microorganisms such as algae, fungi, or bacteria under aerobic or anaerobic conditions. The final line of treatment for produced water is tertiary or chemical treatment which involves the use of chemicals at specified concentrations to treat produced water to make them suitable for various applications[12,4].

Produced water from oil fields contains oil, suspended solids, dissolved solids, traces of radioactive materials, aerobic and anaerobic bacteria as well as heavy metals[8]. Heavy metals in produced water include chromium, copper, iron, zinc, manganese, lead, mercury, cadmium etc at concentrations beyond tolerable limits. Heavy metals are toxic and non-biodegradable and the concentration of heavy metals in produced water depends on the age and geology of the oil and gas reservoir from which the produced water was obtained[8].

The aim of this study is to characterize heavy metals present in produced water obtained from water injection wells in Niger Delta Nigeria, Xray their effects on the ecosystem (humans, plants, animals and environment) as well as the injection wells during enhanced oil recovery. The study is also aimed at assessing the impacts of chemical treatment by hydroxide precipitation on the characteristics of produced water from the injection wells.

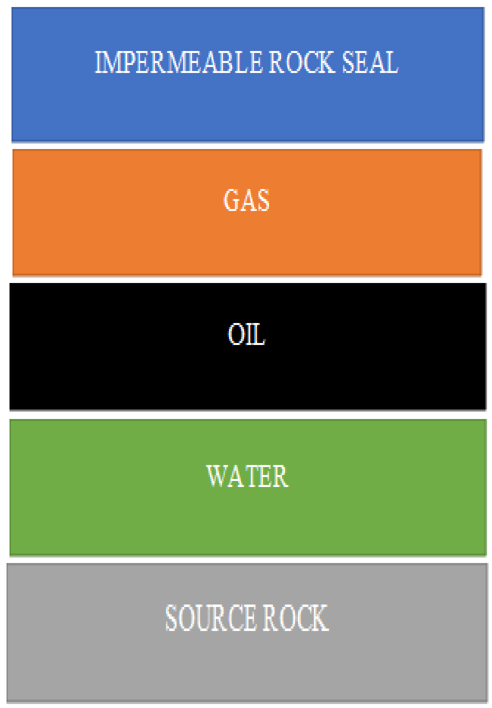

Figure 1: Layers of Reservoir Rock

MATERIALS AND METHODS

Sample Collection / Preparation / Analyses

Produced water from five water injection wells namely KX1, KX2, KX3, KX4 and KX5 in an offshore Location in the Niger Delta area Nigeria were collected respectively using a 1-Litre glass bottle previously cleaned with acetone and dried in an oven. Three samples were obtained per water injection well.

Chemical Treatment of Produced Water by Hydroxide Precipitation

Produced water obtained from the same wells were also treated with different concentrations of $Ca(OH)_2$ as a precipitating agent to get rid of heavy metal in the produced water. Add the required volume of $Ca(OH)_2$ to 1-Litre produced water in a measuring cylinder from each of the wells respectively to achieve the desired concentration. Mix the solution appropriately to obtain a homogenized solution. Allow for a resident time of 24 hours to ensure adequate coagulation of precipitate. Filter water with a membrane filter of 0.5 mm mesh size[8]. Measure the pH and the heavy metal ion concentration of the filtered water. The reaction obtained is represented below:

$$M^{n+} + Ca(OH)_2 \longleftrightarrow M(OH)_n + Ca^{2+} \quad \ldots\ldots(1)$$

Where M is metal

The pH of the filtered produced water was determined using American Society for testing and Materials[13]

Determination of Heavy metal in Produced Water

The heavy metal concentrations in produced water from the water injection wells before and after chemical treatment were determined using Thermo Elemental Flame Atomic Absorption Spectrophotometer (AAS). Samples to be analyzed were digested by adding 2.0 ml of the sample to 4.0 ml of concentrated trioxonitrate v acid (HNO_3) in a 100 ml measuring cylinder and allow to stand for a few hours. Carefully heat the acidified sample over a water bath until the red fumes coming out from the cylinder ceases completely. Allow the cylinder to cool at room temperature and then add 4.0 ml of perchloric acid and then heat the cylinder again over water bath to evaporate to small portion which is filtered through a Whatman filter paper of 0.5mm into another cylinder. Rinse the digesting vessel with distilled water and make up the volume to 100 ml. for analyses. The filtrates were prepared ready for aspiration with the Thermo Elemental Flame Atomic Absorption Spectrophotometer (AAS) using the analytical hollow cathode lamp of interest with the appropriate wavelength and frequency[2]. Analyses were carried out twice with the average heavy metal concentration recorded in mg/l.

RESULTS AND DISCUSSION

Table 1: Heavy Metals and pH of Produced Water before Chemical Treatment

	Water Injection Wells					
Heavy Metal	**KX1**	**KX2**	**KX3**	**KX4**	**KX5**	***EGASPIN RANGE (2022)***
Pb^{2+} (mg/L)	7.558	10.220	8.455	6.268	8.990	0.002 - 8.800
Cd^{2+} (mg/L)	0.200	1.960	1.556	2.000	0.700	<0.005
Cr^{2+} (mg/L)	3.420	4.660	5.220	6.580	2.780	0.020 - 1.100
Ni^{2+} (mg/L)	1.550	1.640	2.800	0.500	0.800	<0.001 - 0.004
As^{2+} (mg/L)	24.220	29.000	13.000	22.000	18.790	0.010 - 35.000
Co^{2+} (mg/L)	0.439	0.425	0.225	0.460	0.900	0.01
V^{2+} (mg/L)	0.083	0.063	0.066	0.100	0.090	0.03
Fe^{2+} (mg/L)	55.890	44.780	49.835	37.890	56.450	<0.100 - 100.000
K^{+} (mg/L)	120.500	98.500	60.807	44.790	1400.000	24.000 - 4300.000
Zn^{2+} (mg/L)	5.600	4.950	3.330	5.050	1.780	<0.010 – 0.700
Hg^{2+} (mg/L)	2.662	2.225	2.970	2.522	2.045	<0.005 - 0.300
Ag^{+} (mg/L)	1.800	1.700	1.850	0.800	1.450	<0.005 - 0.300
Mn^{2+} (mg/L)	65.000	72.000	77.500	60.900	60.500	<0.004 - 175.000
Cu^{2+} (mg/L)	4.860	3.960	4.970	5.240	6.442	<0.02 -1.500
pH	4.800	5.900	6.200	5.100	7.300	4.300 – 10.000

Table 2: Pearson's Correlation for Heavy Metals in Commingled Produced water from five Water Injection Wells with EGASPIN Acceptable limit

Heavy Metal	**Mean(X)**	**Limit (Y)**	**X-M_X**	**Y - M_y**	**$(X - M_x)^2$**	**$(Y - M_y)^2$**	**$(X-M_x)(Y-M_y)$**
Pb^{2+}	8.298	8.800	-28.263	-321.396	798.789	103295.618	9083.579
Cd^{2+}	1.283	0.005	-35.278	-330.191	1244.527	109026.332	11648.444
Cr^{2+}	4.532	1.100	-32.029	-329.096	1025.848	108304.412	10540.58
Ni^{2+}	1.458	0.004	-35.103	-330.192	1232.211	109026.993	11590.695
As^{2+}	21.402	35.000	-15.159	-295.196	229.791	87140.889	4474.839
Co^{2+}	0.49	0.010	-36.071	-330.186	1301.107	109023.03	11910.105
V^{2+}	0.08	0.030	-36.481	-330.166	1330.853	109009.823	12044.752
Fe^{2+}	48.969	100.000	12.408	-230.196	153.962	52990.363	-2856.309
K^{+}	344.919	4300.000	308.358	3969.804	95084.74	15759340.96	1224121.279
Zn^{2+}	4.142	0.700	-32.419	-329.496	1050.982	108567.849	10681.895
Hg^{2+}	2.485	0.300	-34.076	-329.896	1161.164	108831.606	11241.501
Ag^{+}	1.520	0.300	-35.041	-329.896	1227.862	108831.606	11559.851
Mn^{2+}	67.180	175.00	30.619	-155.196	937.532	24085.909	-4751.979
Cu^{2+}	5.094	1.500	-31.467	-328.696	990.163	108041.295	10343.041

Mx: Mean of X = 36.561, My: Mean of Y = 330.196, $\sum(X - Mx)(Y - My) = -1331632.273$, $\sum(X - Mx)^2 = 107769.534$, $\sum(Y-My)^2 = 17005516.691$
Pearson correlation coefficient (R) = -0.9837, Coefficient of determination $(R^2) = 0.9677$

$$R = \frac{\sum(X - Mx)(Y - My)}{\sqrt{\sum(X - Mx)^2 \sum(Y - My)^2}} \quad (1)$$

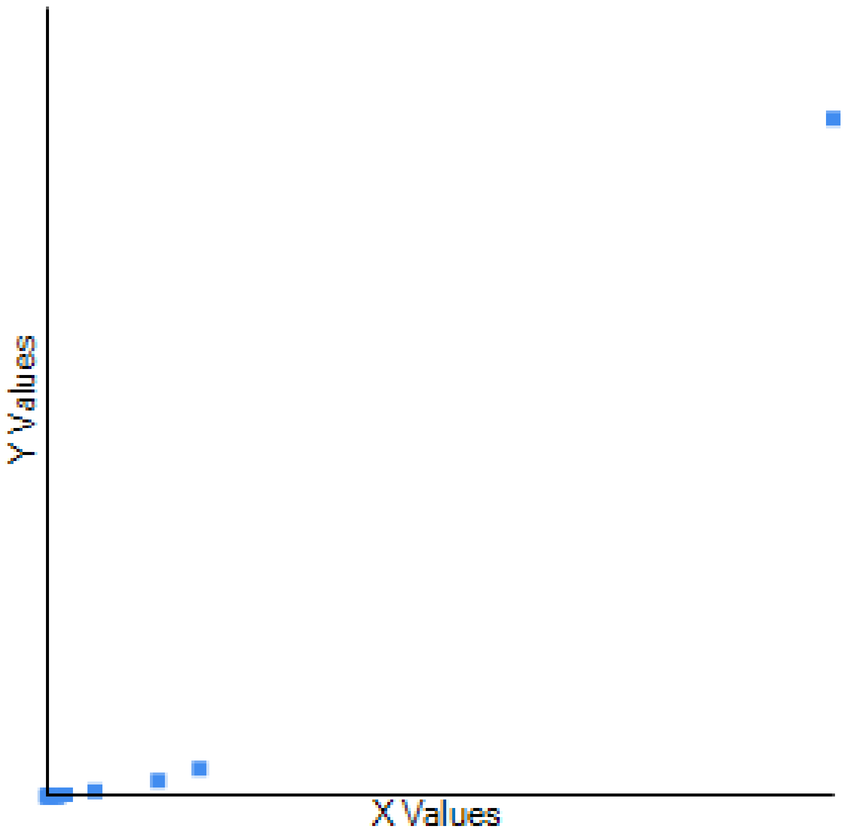

Figure 2: Plot of EGASPIN acceptable limit Vs Mean of Heavy metal concentration

Table 3: Heavy Metals and pH of Produced Water after Chemical Treatment

	Chemical Concentration (ppm)				
Heavy metals	**10**	**20**	**30**	**40**	**EGASPIN Range**
Pb^{2+} (mg/L)	5.8	2.9	1.1	0.5	0.002 - 8.800
Cd^{2+} (mg/L)	0.7	0.05	0.004	0.002	<0.005
Cr^{2+} (mg/L)	2.75	1.25	0.74	0.08	0.020 - 1.100
Ni^{2+} (mg/L)	0.9	0.05	0.007	0.001	<0.001 - 0.004
As^{2+} (mg/L)	15.57	10.45	5.25	1.25	0.010 - 35.000
Co^{2+} (mg/L)	0.4	0.09	0.01	0.005	0.01
V^{2+} (mg/L)	0.05	0.02	0.007	0.003	0.03
Fe^{2+} (mg/L)	35.52	27.48	15.94	8.43	<0.100 - 100.000
K^{+} (mg/L)	285.44	194.6	85.74	44.23	24.000 - 4300.000
Zn^{2+} (mg/L)	2.95	1.45	0.5	0.08	< 0.010 - 0.700
Hg^{2+} (mg/L)	1.99	0.89	0.077	0.008	<0.005 - 0.300
Ag^{+} (mg/L)	1.10	0.5	0.05	0.008	<0.005 - 0.300
Mn^{2+} (mg/L)	48.5	22.5	14.7	8.74	<0.004 - 175.000
Cu^{2+} (mg/L)	3.2	1.8	0.45	0.05	<0.02 -1.500
pH	6.85	7.86	9.01	10.2	4.300 – 10.000

Produced water is an essential part of crude oil exploration considering that they constitute more than half of the produced fluid during drilling operations. Produced water obtained from crude oil contains a considerable concentration of heavy metals which can pose a high degree of hazard to humans, plants and the environment[8]. The proper management of produced water through adequate treatment is essential in guaranteeing the safety of the ecosystem. The mineral bearing rocks forming the crude oil is the source of heavy metals in produced water[1]. The use of produced water as reinjection water into injection wells to boost reservoir pressure and enhance oil recovery is one of the most important uses of produced water in the oil and gas industry[5].

Table 1 shows heavy metal concentrations and pH from five (5) water injection wells before chemical treatment. Results obtained shows that the metal ion concentrations of the heavy metals in all the water injection wells except arsenic (As^{2+}), iron (Fe^{2+}), potassium (K^{+}) and manganese (Mn^{2+}) where above the upper limit of specification stipulated by Environmental Guidelines and Standard for Petroleum Industries in Nigeria (EGASPIN). The concentration of lead (Pb^{2+}) in wells KX1, KX3 and KX4 were narrowly within the upper limit specification.

Table 2 illustrates the Pearson's correlation between the heavy metal ion concentration in the commingled produced water from the five water injection wells and the EGASPIN acceptable limit. Results obtained show a perfectly strong negative correlation (-0.9837) between the metal ion concentrations and the EGASPIN limit which indicates a wide variation between both parameters and this portends a very dangerous trend. This implies that the metal ion concentration in the commingled produced water is far more than what is specified by EGASPIN.

A plot of EGASPIN acceptable limit for each of the heavy metal in the commingled produced water and the average of each metal ion concentration is shown in figure 2. Table 3 shows heavy metal ion concentrations and pH of commingled produced water after treatment with calcium hydroxide. Metal ion concentrations were determined after mixing the produced water with specific concentrations of $Ca(OH)_2$ as shown in Table 3.

The mixing of the water / chemical solution simulates what is obtained from the flow rate of the water / chemical mixture along the expedition line in the Oil / gas process before reception into the wash tank where the commingled produced water is collected. Tanks that receive effluent water also have agitators for proper homogenization of solution after chemical dosage[2]. Hydroxide precipitation is one of the widely used and effective methods for heavy metal ion removal. The hydroxide converts the metal ions dissolved in solution into solid particles for easy sedimentation. $Ca(OH)_2$ precipitates metal ions by changing the pH and electro-oxidizing potential of the solution.

The pH of the produced water before chemical treatment is very important because the chemical precipitates the metal ion by elevating the pH of the solution in fact, sedimentation of metal ions using $Ca(OH)_2$ has proven to be more effective at pH of 9-11[8]. It is therefore important for the pH of the produced water to be reasonably low, possibly within the acidic range to ensure that the pH after treatment is still within EGASPIN specification. Alternatively, carbonate precipitation is more effective at lower pH which makes it necessary for the pH within an alkaline range. Equation 1 shows the reversible reaction between the metal ions in solution and $Ca(OH)_2$ producing a metal hydroxide precipitation with the presence of complexing agents[12]. From Table 3 it can be deduced that the metal ion concentrations of the commingled produced water reduced reasonably after chemical treatment with calcium hydroxide, the higher the chemical concentration, the greater the reduction of the heavy metal ion concentrations. Results also shows that the pH of the commingled produced water increased with increase in chemical concentration, hence the higher the pH the greater the heavy metal ion removal using hydroxide precipitation.

Results obtained from Table 3 shows that treatment with 30 ppm of calcium hydroxide reduced the concentrations of lead (Pb^{2+}), cadmium (Cd^{2+}), chromium (Cr^{2+}), nickel (Ni^{2+}), arsenic (As^{2+}, cobalt (Co^{2+}), vanadium (V^{2+}), iron (Fe^{2+}), potassium (K^{+}), zinc (ZN^{2+}), mercury (Hg^{+}), silver (Ag^{+}), manganese (Mn^{2+}) and copper (Cu^{2+}) within EGASPIN acceptable limit. Higher chemical dosage could further reduce the metal ion which ordinarily would have been an advantage however the relatively large volumes of sludge leading to dewatering, disposal issues as well as the inhabitation of metal hydroxide precipitation is a challenge to contend with[8]. Chemical treatment of produced water must be carried out with the optimal concentration for heavy metal precipitation bearing in mind the concentration of the heavy metal after treatment as well as the volume of sludge recovered[9].

Contaminated and untreated Produced water used as injection water into the reservoir can find their way into groundwater thereby contaminating the water system beneath the earth crust and this is dangerous to humans, plants and animals as groundwater is the primary source of pipe borne water[6]. Heavy metal contaminated ground water can find their way into the soil and then taken up by the plants through their roots. Though heavy metals are insoluble in soil, their solubility is increased by the release of a variety of root exudates which changes their rhizosphere pH thereby making them bioavailable for plant uptake by osmosis[14,7].

Heavy metals introduced into the soil cannot be degraded either biologically or chemically and can persist in the environment for a very long time resulting in environmental pollution and harmful effects to the ecosystem[15]. Heavy metals in plants can exert a variety of toxic actions by damaging plant chloroplast thereby disturbing photosynthesis. Plants occupy a primary place in the food chain for humans and animals, so whatever affects the plants affects humans and animals by extension[7]. Most produced water are used in agriculture for irrigation purposes, exposing agricultural produce to high concentrations of heavy metal can be disastrous both to the agricultural yield, humans and animals that consume these plants[10]. Most produced water from offshore locations are dumped in the sea and this exposes aquatic lives to danger if the heavy metal ion concentration as well as other critical parameters are not within internationally acceptable standards[16]. Heavy metals such as Pb^{2+}, As^{2+}, Mn^{2+}, Ni^{2+}, Cu^{2+} and Zn^{2+} can cause serious health problems to humans. Heavy metals such as Cd and Cr above acceptable limit can cause cardiovascular diseases, kidney related problems, neurocognitive diseases, renal damage and various forms of cancer[17]. High concentration of Cu^{2+} in water can lead to cellular damage resulting in Wilson disease in humans. Exposure to high concentration of As^{2+} either through water and other sources can lead to cardiovascular and peripheral disease, neurologic and neurobehavioral disorders, fibrosis, hematologic disorder etc[18]. Zn^{2+} constitute an important component of human diet however excessive intake of Zn^{2+} above acceptable limit can lead to urinary tract complications resulting in kidney damage, abnormal cramps, diarrhea etc. High concentration of Pb^{2+} in water can lead to liver disease, kidney and heart impairments, memory loss etc[19]. Consumption and assimilation of high concentration of Hg^{+} above permissible limits can lead to impaired deoxyribonucleic acid (DNA) metabolism, genotoxicity and liver damage. Inhalation of V^{2+} vapors can lead to shortness of breath, abdominal pain, greenish discoloration of the tongue while Ni^{2+} ingestion and consumption can lead to cardiovascular disease, lung fibrosis, nasal cancer etc[20]. Consumption of high concentration of silver can cause breathing problems, stomach pain, lung and throat irritation. Excess consumption of potassium (K+) can lead to heart disease, coronary artery disease, hypertension, adrenal insufficiency rate[17].

CONCLUSION

The management of produced water obtained from crude oil exploration is centred on the adequate knowledge of the characteristics of the produced water, this is essential in applying the adequate treatment to ensure proper usage and disposal into the environment. Produced water analysed in this study contains heavy metal ions above acceptable limits. Chemical treatment of the produced water by hydroxide precipitation using adequate chemical concentration of $Ca(OH)_2$ drastically reduced the concentration of the heavy metal by converting the dissolved metal ions to solid particles hence facilitating their sedimentation and subsequent removal from solution by filtration. The resultant produced water after treatment contained heavy metal ions with concentrations that met international standard. Properly treated produced water can be reinjected into the reservoir to enhance oil recovery, used in agriculture for irrigation purposes, discharged into the sea and other water bodies during offshore operations or even used in drilling services. Heavy metal ions in produced water can find their way into groundwater after reinjection thereby contaminating pipe borne water, they can also find their way into the soil, taken up by the plants through their roots. Heavy metals in plants can exert a variety of toxic actions by damaging plant chloroplast thereby disturbing photosynthesis. Plants occupy a primary place in the food chain for humans and animals, so whatever affects the plants affects humans and animals by extension. Consumption, ingestion and inhalation of heavy metals by humans can cause a wide range of ailments such as cardiovascular diseases, kidney related problems, neurocognitive diseases, renal damage, heart disease, coronary artery disease, lung fibrosis, nasal cancer etc

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