# HEAVY METAL HUMAN HEALTH RISK ASSESSMENT OF STABILIZED/SOLIDIFIED LOW-TEMPERATURE THERMALLY DESORBED OIL-BASED DRILL CUTTINGS

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## ABSTRACT

Drill Cuttings are the various rock particles and liquids released in the drill hole during oil and gas exploration. When drill cutting is stabilized/solidified, it can be reused as construction materials. Consequently, there is the need for risk assessment for the human health risk of reused stabilized/solidified drill cutting as to make good and environmentally safe decision for reuse. The Stabilized/Solidified drill cuttings was cast in different grades of concrete, cured for 28days, then dried and grinded before laboratory analysis for heavy metal. Evidently, the different grades treatments shows that the completely Randomized Design One Way ANOVA is highly significant with concrete grades 10, 15, 20 and 25 of heavy metals. The contamination indices like contaminant factor indicate suspicion of heavy metal presence while Pollution Load Index is 5.31 showing that heavy metal pollution exists. The heavy metal in the Stabilized/Solidified drill cutting was assessed basically in the three exposure pathway for human via ingestion of chemical in the soil, ingestion in drinking water and dermal contact with the soil in the assessed heavy metal. The Hazard Index of Barium 1.20E-1, Zinc 1.6E0 and Lead 1.2E-1, while Arsenic and Cadmium are negligible implying that the stabilized/solidified drill cutting has no adverse effect on the residence since the hazard index is less than 1, except zinc. The heavy metal (Lead) by oral access has a total risk of 3.72E-6 implying a total lifetime cancer risk of an approximate 4 adult persons out of **1**Million persons in Niger Delta Therefore, it is still not advisable to reuse drill cuttings for construction of residential facilities after it treatment rather risk assessment should be conducted.

**Keywords:** Drill cutting; heavy metal; risk; hazard index and cancer risk.

## INTRODUCTION

Heavy metals existed in the environment through natural process and human activities (Abdullah et al; 2011). The natural sources of heavy metal gained access to the environment via acidification, erosion and weathering process (Tamjam and Kamal 2013). Human activities (anthropogenic sources) such as industrial processes, domestic wastes, agricultural activities and emissions from vehicles and factory plants are the main sources for some heavy metals entering and deposition into the environment (Demirak 2013). Heavy metals are well known to be toxic to most organisms and humans in the environment when present in excessive concentrations (Giller, 1998; Hsiau et al., 1998 Okparanma et al., 2009).

Heavy metal toxicity can reduce energy levels and cause damage to the brain, lungs, kidney, liver, blood composition and other important organs. Long-term exposure can lead to multiple sclerosis, Parkinson's disease, Alzheimer's disease, muscular dystrophy and cancer (Jarup, 2003, Ayotamuno et al., 2007 and Koki et al., 2015).

## LITERATURE REVIEW

### Stabilization/Solidification (S/S) of Drill Cuttings

Stabilization/solidification (S/S) is one of the major methods in treating hazardous wastes prior to reusing of DC, and also an effective technique for reducing the leachability of contaminant (heavy metal) in soils (Gupta, 2007; Kogbara et al., 2011 Hytiris et al., 2014). The entrapment of wastes that express hazardous characteristics within a cementitious matrix (solidification) and the binding of the contaminants (organic or inorganic) of a hazardous stream into a stable insoluble form (stabilization) are the mechanisms that best describe the principle behind solidification and stabilization (S/S) treatment.

The most commonly used primary binder for S/S matrix is Portland cement because it can restrict the mobility of heavy metals due to high pH and due to its capability to precipitate the metals in insoluble forms (Gupta, 2007; Kogbara et al., 201, and Okparanma and Ayotamuno (2008).

### Leachability of Stabilized/Solidified Drill CUTTINGS

Solidification/Stabilization of drill cuttings for construction will mostly be in contact with the ground. Considering the Niger Delta terrain with low water table for about a period of 70 years will possibly cause leaching of the entrapped heavy metal in the drill cuttings. The usual receptor to leaching is water bodies (surface and groundwater) and the soil. This is clear indication of the pathway to the environment and human's inclusive. Water as a universal solvent in contact with stabilized/solidified DC will inevitably lead to changes in the chemistry and these changes may be affecting the stability of the waste (DC) as shown in the conceptual model in figure 1

## METHODOLOGY

### Study Area

The Niger Delta is located in the Atlantic coast of Southern Nigeria and is the world's second largest delta with a coastline of about 450km which ends at Imo river entrance (Awosika, 1995) During 1991 National Census estimation of about 25% of the entire Nigerian population lives within the Niger Delta region (Twumasi and Merem, 2006; Uyigue and Agho, 2007).

### Sample Preparation

About 1g of sample was weighed and put in a 500ml round bottom flask. A 100ml of distilled water was added. Thereafter 2ml of (1 + 1) nitric acid and 10ml of (1+ 1) HCl acid was added. Swirled and placed on a heating mantle, heat until the volume reduce to about 15ml to 25ml, allow to cool and then filter

### Heavy Metal Analysis

The heavy metal analysis was done with the help of PG Instruments – AA500 Spectrophotometer.
The metals have different wavelengths; the equipment is calibrated using a specify standard and lamp unique for each of the metals and a linear graph is generated prior to analyzing the metals of interest.
The concentrations of heavy metals in the extract produced by toxicity characteristic leaching procedure (TCLP) were determined by digestion with HF-HNO3-HClO4, the concentration was determined by flame atomic absorption spectrometry (AAS). The reading was taken with the computer attached to the PG Instrument.

### Casting and Curing of the Concrete

A binding material (Portland cement) was mixed together with sand, coarse aggregate and water in a water-dry binder ratio (COREN 2017) and (Anum et al., 2014), for Grade 10 is 1:4:8:0.95. For Grade 15 is 1:3:6:0.8 for Grade 20 is 1:2.5:5:0.7. For Grade 25 is 1:2:4:0.6 and the sand were replaced with DC at 4%, 6%, 8% and 10%. After thorough mixing, the mixture was cast into cylindrical moulds of $0.15m^3$ and allowed to cure for 28 days in an open water tank.

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The different structural grade was to enable the observation of any fractional variation in the concentration of the chemical in the DC. While, the water, coarse aggregate and cement quantity remain constant, the sand and DC is varied according to percentages added. Thus increase in the DC decreases the sand content of the concrete grade.

Table1 below shows the replacement variation done to the concrete, the samples from each mix was tested for their toxicity characteristic leaching procedure.

Table 1:Percentages of Replacement of Sand With DC

CONCRETE GRADE (G10)

| Constituent | 4% | 6% | 8% | 10% |
|---|---|---|---|---|
| Cement | 0.72 | 0.72 | 0.72 | 0.72 |
| Sand | 2.76 | 2.70 | 2.64 | 2.58 |
| Drill cutting | 0.12 | 0.17 | 0.23 | 0.28 |
| Aggregate | 5.73 | 5.73 | 5.73 | 5.73 |
| Water | 0.68 | 0.68 | 0.68 | 0.68 |

CONCRETE GRADE (G15)

| Constituent | 4% | 6% | 8% | 10% |
|---|---|---|---|---|
| Cement | 0.92 | 0.92 | 0.92 | 0.92 |
| Sand | 2.67 | 2.61 | 2.56 | 2.50 |
| Drill cutting | 0.11 | 0.17 | 0.22 | 0.28 |
| Aggregate | 5.56 | 5.56 | 5.56 | 5.56 |
| Water | 0.74 | 0.74 | 0.74 | 0.74 |

CONCRETE GRADE (G20)

| Constituent | 4% | 6% | 8% | 10% |
|---|---|---|---|---|
| Cement | 1.10 | 1.10 | 1.10 | 1.10 |
| Sand | 2.61 | 2.56 | 2.50 | 2.45 |
| Drill cutting | 0.11 | 0.16 | 0.22 | 1.27 |
| Aggregate | 5.43 | 5.43 | 5.43 | 5.43 |
| Water | 0.75 | 0.75 | 0.75 | 0.75 |

CONCRETE GRADE (G25)

| Constituent | 4% | 6% | 8% | 10% |
|---|---|---|---|---|
| Cement | 1.31 | 1.31 | 1.31 | 1.31 |
| Sand | 2.51 | 2.46 | 2.40 | 2.35 |
| Drill cutting | 0.10 | 0.16 | 0.21 | 0.26 |
| Aggregate | 5.23 | 5.23 | 5.23 | 5.23 |
| Water | 0.85 | 0.85 | 0.85 | 0.85 |

### Determination of Contaminant Indices of Heavy Metal

The heavy metal and the contaminant indices used in the analysis besides other contaminant index includes; Contaminant Factor (CF) and Pollution Load Index (PLI).

### Contaminant Factor (Cf)

The parameter CF was determined to express the level of heavy metals (Ba. Pd. and Zn) contamination in drill cuttings treated with S/S. The CF was calculated using equation 1.

$$CF = \frac{C_{Metal}}{C_{Background}} \quad (1)$$

**Pollution Load Index (Pli)**

The PLI of Ba, Pd and Zn was evaluated using the procedure by Tomlinson et al., (1980), according to equation 2.

$$PLI = (CF_1 \times CF_2 \times CF_3)^{1/n} \quad (2)$$

Where: n = number of metals and CF = Contamination Factor.

The C.F were then classified using Table2 developed by Tomlinson et al., (1980)

Table2 :Contaminant Factor Scale

| C.F Value Scale | Classification |
|---|---|
| 1 and less | No contamination |
| 1-2 | Suspected |
| 2-3.5 | Slight |
| 3.5-8 | Moderate |
| 8-27 | Severe |
| 27 and above | Extreme |

The values of equations 1 and 2 are scaled by Table2.

**Heavy Metal Exposure Assessment Quantification**

For dermal contact with soil, the allowable daily dose intake (ADI) was calculated with equation 3

$$ADI = \frac{CS \times CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (3)$$

For ingestion of chemical in soil the chronic daily intake (CDI) was calculated using equation 4

$$CDI = \frac{CS \times IR \times CF \times FI \times EF \times ED}{BW \times AT} \quad (4)$$

For ingestion in drinking water, the CDI was deduced using equation 5

$$CDI = \frac{CW \times IR \times EF \times ED \times DAF}{BW \times AT} \quad (5)$$

The necessary parameters to be substituted into equations 3, 4 and 5 are shown in Table3

Table3 : Exposure Standard Parameters

| Symbol | Parameter=Standard Value | References |
|---|---|---|
| CS | Chemical Concentration in Soil=specific chemical | |
| CF | Conversion Factor = 0.000001 kg/mg | USEPA 1989 |
| SA | Skin Surface Area Available for Contact= $5700 cm^2$ | USEPA. 2004 |
| AF | Soil -to-Skin Adherence Factor=$0.07 mg/cm^2$ | USEPA 1992, |
| ABS | Absorption Factor for Soil Contaminant=0.15 unitless | USEPA 1989 |
| EF | Exposure Frequency, Residential= 350 day/year | USEPA 1991 |
| ED | Exposure Duration, Adult Resident=70years | USEPA 1989 |
| IR | Soil Ingestion, Adult= 100 mg/day,2L/day | USEPA 1991 |
| FI | Fraction Ingestion=0.5unitless | USEPA 1989 |
| DAF | Dilution Attenuation Factor = 1 | EPA, 1996 |
| BW | Body Weight, Adult= 70 kg | USEPA 1989 |
| AT | Averaging Time=ED$\times$ 365 = 25,550 day | USEPA 1989 |
| CW | Chemical Concentration in Water=Specific chemical | |

**Exposure Hazard Assessment**

Hazard quotient, HQ, was calculated using equation 6

$$HQ = \frac{ChronicDailyIntake(CDI)\ (mg/kg.day)}{ReferenceDose\ (mg/kg.day)} \quad (6)$$

The total chronic hazard attributable to exposure to all COPCs through a single exposure pathway is known as a Hazard Index (HI). The HI was calculated using equation 7:

HI = $\sum$ HQi (7)
Where,
i=increment
HI = Hazard Index for a specific exposure pathway
HQi= Hazard Quotient for COPC i
HQ<1=Safe while HQ>1=Unsafe

**Exposure Risk Assessment**
To quantify the amount, the expression in equation 8 or 9 was adopted according to USEPA (1989)

$$\text{Risk}=\text{CDI (mg/kg. day) xCancerPotential Factor (CPF)}\left(\frac{mg}{kg.day}\right)^{-1} \quad (8)$$

Or

$$\text{Risk}=\text{ADI (mg/kg. day) xCancerPotential Factor (CPF)}\left(\frac{mg}{kg.day}\right)^{-1} \quad (9)$$

The necessary parameters substituted into equations 8 and 9 are contained in Table4

Table4 : Toxicity Assessment Parameters

| S/N | Heavy Metal | ORAL RfD (mg/kg.d) | DERMAL RfD (mg/kg.d) | CPF$\left(\frac{mg}{kg.day}\right)^{-1}$ | References |
|---|---|---|---|---|---|
| 1 | Barium | 7.00E-2 | | | USEPA 2004 |
| 2 | Zinc | 3.00E-2 | 7.50E-2 | | USEPA 2004 |
| 3 | Lead | 3.60E-3 | | 8.50E-3 | USEPA 2004 |

**Estimation of Cancer Risk**
Total lifetime cancer risk was deduced with equation 10
Total Lifetime Cancer Risk (TLCR)= Risk × Population (10)

**RESULT**
The conceptual model figure1 explains the the leachability of the heavy metal entrapped in the stabilized/solidified drill cuttings reused for construction purposes.

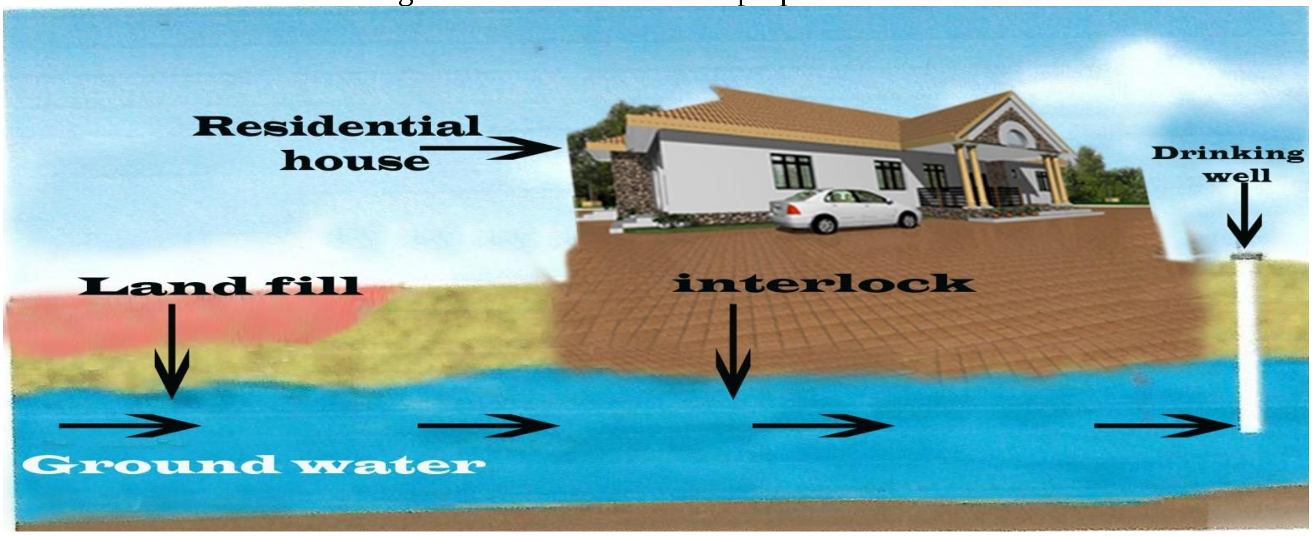

Figure1: Conceptual model of Heavy Metal leachabilty

Table5: Pre-treated Drill Cuttings Concentration

| Heavy Metal | concentration (mg/kg) | DPR2002 limit (mg/kg) |
|---|---|---|
| Arsenic | < 0.01 | 5 |
| Barium | 0.42 | 100 |
| Cadmium | < 0.01 | 1 |
| Lead | 0.027 | 5 |
| Zinc | 29.12 | 50 |

< Limit of Quantification is 0.01

Table6: Contamination Factor (C.F)

| Heavy Metal | Concrete Grade | Treated Mean | C.F | Pre-treated Mean | C.F |
|---|---|---|---|---|---|
| Barium | $G_{10}$ 4% | 0.32 | 1.14 | 0.42 | 1.5 |
| Lead | $G_{10}$ 4% | 0.018 | 1.33 | 0.027 | 2.7 |
| Zinc | $G_{10}$ 4% | 18.128 | 1.06 | 29.12 | 1.71 |
| Barium | $G_{15}$ 6% | 0.28 | 1.4 | 0.42 | 2.1 |
| Lead | $G_{15}$ 6% | 0.014 | 1.58 | 0.027 | 3 |
| Zinc | $G_{15}$ 6% | 17.13 | 1.27 | 29.12 | 2.15 |
| Barium | $G_{20}$ 8% | 0.26 | 1.01 | 0.42 | 1.62 |
| Lead` | $G_{20}$ 8% | 0.016 | 1.81 | 0.027 | 3 |
| Zinc | $G_{20}$ 8% | 15.515 | 1.17 | 29.12 | 2.2 |
| Barium | $G_{25}$ 10% | 0.341 | 1.36 | 0.42 | 1.68 |
| Lead | $G_{25}$ 10% | 0.021 | 1.71 | 0.027 | 2.25 |
| Zinc | $G_{25}$ 10% | 18.79 | 1.34 | 29.12 | 2.08 |
| Conclusion | | Suspected Contamination | | Slight and Suspected Contamination | |

Table7: Pollution Load Index (PLI)

| Conrete Grade | Pre-treated | Treated |
|---|---|---|
| G10 4% | 1.91 | 1.7 |
| G15 6% | 2.38 | 1.39 |
| G20 8% | 2.20 | 1.29 |
| G25 10% | 1.99 | 1.46 |
| | $\sum$ 8.478 | $\sum$ 5.31 |

Table8: Receptor Route Quantification

| | Pre-treated DC | | | Treated DC | | |
|---|---|---|---|---|---|---|
| Heavy metal | ADI | $CDI_S$ | $CDI_W$ | ADI | $CDI_S$ | $CDI_W$ |
| Barium | 2.79E-7 | 2.88E-7 | 1.15E-2 | 2.13E-7 | 2.06E-7 | 8.23E-3 |
| Zinc | 2.07E-5 | 1.99E-5 | 7.98E-1 | 1.24E-5 | 1.19E-5 | 4.8OE-1 |
| Lead | 1.92E-8 | 1.85E-8 | 7.40E-4 | 1.14E-8 | 1.10E-8 | 4.38E-4 |

ADI= *AbsorbedDailyDose Intake*e
$CDI_S$ = Chronic Daily Intake via Soil
$CDI_W$= Chronic Daily Intake via Water

Table9: Hazard Quotient and Index

| Heavy metal | $HQ_A$ | $HQ_S$ | $HQ_W$ | HI |
|---|---|---|---|---|
| Barium | 3.04E-6 | 2.94E-6 | 1.20E-1 | 1.2E-1 |
| Zinc | 1.65E-4 | 3.97E-5 | 1.60E0 | 1.6E0 |
| Lead | NA | 3.10E-6 | 1.20E-1 | 1.2E-1 |

NA=Not Availabe
$HQ_A$=Hazard Quotient for Dermal Contact with Soil
$HQ_S$=Hazard Quotient for Soil Ingestion
$HQ_W$=Hazard Quotient for Ingestion via Drinking Water
HI=Hazard Index

Table10: Risk In Drill Cuttings

| Heavy metal | $RISK_S$ | $RISK_W$ | $RISK_T$ |
|---|---|---|---|
| Lead | 9.35E-11 | 3.72E-6 | 3.72E-6 |

$RISK_S$= Risk of Soil Ingestion
$RISK_W$=Risk of ingestion via drinking water
$RISK_T$= Total Risk

## DISCUSION

These value in Table5 were below the DPR (2002) limit, however, there is need to ascertain the probability of risk of reuse even after S/S treatment

### Application of Contamination Factor

The CF of Ba, Pd, and Zn in the DC varies differently at each grade of the concrete as contained table6, however no significant variation in the percentage of replacement of sand with DC, but between G7 and G20 of the concrete, there is an appreciable increase in heavy metal. There is suspected contamination of DC with heavy metal, which is lower than the contamination status before the S/S treatment, before treatment with S/S, the contamination factor was between suspected to slight contamination.

### Application of Pollution Load Index (PLI)

PLI is a potent tool in HM pollution evaluation (Hankanson L 1980 and Tomlinson et al., 1980) PLI >1 indicates that pollution exist while PLI < 1 indicates no pollution occurred. The PLI of Ba, Pd and Zn also varies at each concrete grades. The PLI values of heavy metal in the DC are contained in the table 7. It shows the before and after S/S treatment with respect to the control sample at various grades. Judging from table7, PLI>1, meaning that pollution exist in stabilized/solidified drill cuttings. Stabilization/Solidification of drill cuttings with the laboratory result shows that the mean value of contaminant in the different grades in table 7 indicating a definite reduction in the concentration.

The experimental design technique for this research work is completely randomized design (CRD). The Computed $F_{Value}$ is larger than the Tabular $F_{Value}$ that means highly significant. The Coefficient of Variation (CV) indicates high reliability of the experiment (Zady, 1999). It also shows that all observed difference amongst the percentile increase and replacement treatment is very small. Furthermore since the treatment means is far greater than the error means; it shows that, the changes in the concentration of the heavy metal are due to the treatment variation of the DC.

**Hazard and Risk Assessment**

**Dermal Contact with Soil**

Consequent upon the leaching of the drill cuttings to the soil, contaminant absorption through dermal contact may contribute risk to human health in a residential setting.

**Ingestion of Chemical in Soil**

The chronic exposure to non carcinogens and carcinogens through direct ingestion of contaminated soil in a residential setting can contribute risk the environment and human.

**Ingestion of Chemical in Drinking Water**

The drill cuttings leachate can move through soil to ground water since it is use for construction of structure that must inevitably touch water, the contaminant concentrations will be attenuated by adsorption and degradation EPA (2002b). This reduction in concentration can be expressed by a dilution attenuation factor (DAF).DAF was defined by EPA, 1996 and DEQ, 2018 as the ratio of stabilized/solidified concentration migration to receptor location of concentration. The lowest possible DAF is 1, which is an equivalence of the concentration in the receptor well and the stabilized/solidified concentration

**Hazard Assessment**

The Receptor Route for each chemical contaminant in the DC is shown in Table8. The result in Table9 shows that ($HQ_W$) ingestion of chemical in drinking water is far higher than other routes of assessment, therefore drinking water that is contaminated with heavy metal is dangerous.

After calculating the total chronic hazard for each exposure pathway by following the procedures outlined in USEPA (2018), the results in table9, indicated that HI values for Barium and Lead assessed through both ingestion and dermal adsorption is less than 1, while the HI of Zinc is greater than 1 indicating danger of reuse of DC for construction.

**Risk Assessment**

Lead at extremely low concentrations, are toxic and can cause many diseases, including the increased risk of Cancer (Willers et al., 2005 and Yan et al., 2012). Lead carcinogenicity is evaluated via oral assess by ingestion of soil and drinking water. In table 10 the risk value in drinking water shows that it is cancerous

**Total Lifetime Cancer Risk (TLCR)**

The Niger Delta region has a steady growing population of approximately 30 million people as of 2005, accounting for more than 23% of Nigeria's total population Twumasi and Merem, 2006; Uyigue and Agho 2007).In determining the lifetime cancer risk of exposure to stabilized/solidified drill cuttings reused for construction purposes.Thus, TLCR for Lead contamination after oral source route shows that about 4 adult persons out of 1Million of people in Niger Delta will be in danger.

**CONCLUSION**

The contamination from heavy metal is dangerous to the environment. The S/S treatment of DC helps in reducing the leachability of heavy metal in the environment; however, regular contact with water will obviously affect the heavy metal content. The CF after the S/S treatment is suspected contamination, and the PLI is 5.31 indicating that the DC is polluted, hence, the need to evaluate the carcinogenicity and non- carcinogenicity. The knowledge of these two contamination indices are the basic key needed in decision making about the reuse of DC for construction.

In the three route considered; dermal contact with soil, soil ingestion and ingestion via water, the heavy metal contamination is more in the ingestion via water drinking than any other route especially the Zinc. Amazingly, after S/S DC, it shows that the DC is contaminated, although non-carcinogenic but the lead is risky and it's carcinogenic. It shows that 4 persons in Niger Delta out of 1million can have cancer. In the x-rayed route, it shows that leaching into water contributes very high amount of contamination. Therefore, the drinking of water exposed to stabilized/solidified DC should be avoided.

**Conflict of Interest**

The authors declare that there is no financial or commercial conflict of interest.

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