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## Remediation of Some Heavy Metals, Radionuclides and Bacterial Load of Medically Acclaimed Edible Kaolin

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ARTICLE INFO	ABSTRACT
Article history: Received 20 March 2018 Revised 05 May 2018 Accepted 06 May 2018 Published online 10 May 2018	The earliest recorded mention of clay consumption for healing remedy originated approximately 5,000 years ago. But edible clays have been implicated to contain microbial and metallic contaminants despite the inherent medical benefits. This work reports the remediation of some heavy metals, radionuclides and bacterial load from edible kaolin from Kankara, Nigeria. The remediation increased the pH of the samples except when HCl was used; though the colour and taste remained unaltered. The best extractant for arsenic in the kaolin sample was 0.1M HCl with

**Copyright:** © 2018 Omoniyi *et al.* This is an openaccess article distributed under the terms of the <u>Creative Commons</u> Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited. The callect recorded method of cary consumption for neuron for neuron originated approximately 5,000 years ago. But edible clays have been implicated to contain microbial and metallic contaminants despite the inherent medical benefits. This work reports the remediation of some heavy metals, radionuclides and bacterial load from edible kaolin from Kankara, Nigeria. The remediation increased the pH of the samples except when HCl was used; though the colour and taste remained unaltered. The best extractant for arsenic in the kaolin sample was 0.1M HCl with efficiency of 19%, H<sub>2</sub>O with efficiencies of 98% and 75% was best for removal of Cr and Pb respectively. The extraction resulted to significant reduction of the concentrations of Cr and Pb in the clay(P < 0.05). The remediation of <sup>232</sup>Th and <sup>238</sup>U was best by using surfactant solution (Sodium dodecyl sulphate, SDS)with efficiencies of 46% and 35% respectively; while 0.1M CaCl<sub>2</sub> with 17% efficiency was the best for<sup>228</sup>Ra removal. All the clay samples were contaminated with bacteria. Therefore, soil washing technique can be employed to reduce contaminants in edible clay while maintaining the inherent medical benefits of geophagy.

Keywords: Edible Kaolin, Extractant, Kankara, Contaminants, Geophagy.

## Introduction

The earliest recorded mention of clay consumption for healing remedy was the healing tradition of India that originated approximately 5,000 years ago.<sup>1</sup>Clay consumption is prompted by pregnancy to eliminate nausea,<sup>2</sup> absorb dangerous toxins in the body,<sup>3</sup> so as to born beautiful children,<sup>4</sup> have lighter and softer skin,<sup>1</sup> and a plethora of others.

Since the act of eating the earth (known as geophagia) including clay and chalk is neither new nor outdated;<sup>5</sup> several research have focused on the assessment of several contaminants in geophagious clay. These include the presence of 22 elements in calabash chalk ('Nzu clay'), including lead, chromium, arsenic, zinc among others.<sup>6,7,8,9</sup> It has also been reposted to contain microbes like bacteria – *Bacillus subtilis*, Staphylococcus *aureus*, *Escherichia coli*, Klebsiellae *and* Helminthes – *Ascaris lumbricodies* and hookworm<sup>10</sup> as well as radionuclides - <sup>90</sup>Sr, <sup>134</sup>Cs, <sup>137</sup>Cs, <sup>238</sup>Pu, <sup>239+240</sup>Pu, <sup>241</sup>Am, <sup>242</sup>Cm, <sup>234</sup>Th, <sup>235</sup>U, <sup>40</sup>K,<sup>11,12</sup>

<sup>134</sup>Cs, <sup>137</sup>Cs, <sup>238</sup>Pu, <sup>239+240</sup>Pu, <sup>241</sup>Am, <sup>242</sup>Cm, <sup>234</sup>Th, <sup>235</sup>U, <sup>40</sup>K.<sup>11,12</sup> Edible kaolin is called 'Farar kasa' (Hausa), 'Ndom' (Efik/Ibibio, Nigeria), 'Shilè' in Ghana,<sup>10,13</sup> despite the reported health implications of consuming clay, its addiction has been established across ages, gender and races.<sup>13</sup>Since small amounts of heavy metals and radioactive elements occur naturally in rocks, clay and soil particles. The concentrations of which depend on anthropogenic processes, geology of the environment and other natural processes.<sup>14</sup>The concentrations of some of these elements in soils are reported thus: uranium 2 – 3 mg/L, thorium 8 – 12 mg/L, potassium 0.1% for sandstone, 1% sandstone and 3.5% for granite.<sup>15</sup>Also, activity concentration of <sup>238</sup>U in standard soil

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has a mean of 35 Bq/kg and range of 17- 60 Bq/kg, for  $^{232}$ Th the mean is 30 Bq/kg and range of 11- 64 Bq/kg.<sup>16</sup> The reported activity concentration of  $^{226}$ Ra in soil/rock range from 19.2  $\pm$  5.6 Bqkg<sup>-1</sup> to 94.2  $\pm$  7.7 Bqkg<sup>-1</sup> with mean of 41.0  $\pm$  5.0 Bqkg<sup>-1</sup>.<sup>10, 17</sup>

Therefore, the removal of pollutants from environmental media – soil, groundwater, sediment, surface water – is termed remediation. Soil washing is one of the techniques used, it is time-efficient and versatile.<sup>18</sup> Water, 1M NH<sub>4</sub>Cl, 0.01M CaCl<sub>2</sub>, 0.005M, diethylenetriamine pentaacetice acid (DTPA), 0.1M ethylenediamine tetraacetic acid (EDTA), 0.1M HCl and 1M HCl to remediate cadmium, zinc and lead from soil.<sup>19</sup>Also, the mechanism of removal of metals from contaminated soils was done using water and surfactants,<sup>20</sup> humic acid.<sup>21</sup>

The aim of this work is to remove some heavy metals, radionuclides and microbes from edible kaolin, so as get rid of the implicated contaminants and maintain the medical benefits of edible kaolin to the consumers.

## **Materials and Methods**

## Sample collection

Five raw edible kaolin samples were collected from Farar Kasa hills in Kankara, Kankara Local Government Area of Katsina State, Nigeria and were coded EKK. Also, five processed samples of Farar kasa (whose source was traced to Kankara, Nigeria) were purchased from Sabo and Samaru markets in Zaria, Kaduna State, Nigeria and coded EKM (n = 10), presented in Plate 1. Then each set of EKK and EKM were pooled into a composite.

## Experimental setup and operation

Each set of the samples (EKK and EKM) were air-dried at ambient temperature  $(28 - 31^{\circ}C)$  and pulverized using a porcelain mortar and pestle. Each experimental group was sieved using 2.00 mm sieve, after which, 500 g of each sample was weighed, mixed thoroughly and then aggregately selected for experimental tests.

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Plate 1: Processed edible kaolin (Farar kasa) samples.

#### Determination of organoleptic properties and pH

The organoleptic properties (colour and taste) and pH of each clay sample (EKK and EKM) were determined to ascertain the effect of the remediating solution on the clay samples before and after remediation. The pH of each clay sample (EKK and EKM) was determined by weighing 15 g of each clay sample into a 100 mL beaker. This was suspended in 30 mL of a 0.01M CaCl<sub>2</sub> and agitated for 5 minutes, and then allowed to settle for 30 min. The pH was measured at room temperature using a Jenway pH meter (model HI 93710, Switzerland). The colour of each clay sample was determined by using a 10% clay:water mixture, and was shaken for 1 h in a rotary shaker. The resulting suspension was allowed to stand for 24 h in order to settle. The resulting solution was decanted and the colour was determined using a Lovibond 1000 colour comparator. The Hazan NSA disc was used to assess the colour.

A panel of 15 analysts was used to test for the taste of each clay sample (EKK and EKM) before and after remediation. Their various observations were noted and recorded appropriately.

#### Determination of Heavy Metals

In order to determine the concentration of heavy metals in each of the clay samples (EKK and EKM). Wet digestion was carried out on 1.0 g of each clay sample using aqua regia (1:3 of ultra-pure 70% HNO<sub>3</sub> and pure 37% HCl). After digestion, each of the digest was filtered using a Whatman number 42 filter paper. The filtrate was analysed for heavy metals using a Microwave Plasma Atomic Emission Spectrophotometer at the Multi–user Laboratory, Ahmadu Bello University, Zaria – Nigeria. The total extractable Cd was determined at 228.802 nm, Pb at 405.781 nm, Cr at 425.433 nm and As at 193.695 nm.

The removal efficiency of the heavy metals was calculated using the following equation:  $^{\rm 22}$ 

RemovalEfficiency (%) = 
$$\frac{C_0 - C_f}{C_0} \times 100$$

Where,  $C_0$  is the initial concentration of the metal in each clay sample and  $C_f$  is the final concentration of metals after remediation of the clay samples.

## Determination of radionuclides

Similarly, the activity concentrations of <sup>228</sup>Ra, <sup>238</sup>U and <sup>232</sup>Th in each of the kaolin samples (EKM) were determined using High Purity Germanium detector (HPGe) at the National Institute of Radiation Protection and Research, University of Ibadan, Ibadan – Nigeria in 2017.

#### Determination of microbial load

The method for determining the microbial load in the EKK and EKM samples was reaffirmed at the Department of Microbiology, Ahmadu Bello University, Zaria. This was done by mixing the clay sample (25 g) in 100 mL of sterile saline solution for 2 min under sterile conditions. The homogenates were then collected in sterile bottles and stored at -20°C until needed. Aliquots (0.5 mL) of each homogenate were serially diluted in sterile saline solution. The diluent of buffered peptone water was then inoculated on to the respective media. *E. coli* in each edible clay was determined by the procedure described by IS 5887(I):1976.<sup>23</sup>

## Remediation Procedures

Remediation was done using distilled water, 0.1M HCl, 0.1M CaCl<sub>2</sub> and surfactant solution (Sodium Dodecyl Sulphate, SDS and Triton).

## Remediation using distilled water

For each of the EKK and EKM clay samples, 10 g was suspended in a 120 mL polypropylene bottle, then 100 mL of distilled water was added. The bottle with its content was agitated for a contact time of 1 h in a Heldoph rotary shaker.<sup>19</sup> Then all suspensions were centrifuged in a centrifuge machine for 10 min at 10000 rpm, the residues were then obtained by decantation.<sup>21</sup> The residues were air-dried at room temperature and the dried clay samples were then pulverized using an agate mortar and pestle. The particles were then sieved and kept in a desiccator prior to the determination of the level of heavy metal, radionuclide and microbe. This washing removes the water-soluble contaminants.<sup>19</sup>

## Remediation using 0.1M calcium chloride

For each of the EKK and EKM clay samples, 10 g was suspended in a 120 mL polypropylene bottle, then 50 mL of 0.1M CaCl<sub>2</sub> was added. The bottle with its content was agitated for a contact time of 16 h in a Heldoph rotary shaker.<sup>19</sup> Then all suspensions were centrifuged in centrifuge machine for 10 minutes at 10000 rpm. The air-dried solid residues were pulverised and kept in a desiccator prior to heavy metal, radionuclide and microbe determination.<sup>21</sup> This washing removes the exchangeable/neutral salt soluble contaminants.<sup>19,24</sup>

## Remediation using 0.1M HCl

Ten gramme of each of the edible kaolin clay suspended in 100 mL of 0.1M HCl solution was shaken for 30 min in a rotary shaker.<sup>19</sup> Then the suspension was centrifuged in glass tubes at for 10 min in order to separate solid residues from the washing solutions. The solid residues were dried at 40°C.<sup>21</sup> The dried clay samples were crushed and prepared for determination of heavy metals, radionuclides and microbes. This washing was targeted at removing the weak acid extractable contaminants.<sup>19,25</sup>

#### Remediation using surfactant

This was done by placing 10 g of each of the clay samples (EKK and EKM) in a 120 cm<sup>3</sup> polypropylene bottle, then 100 mL of surfactant solution was added (this was made up of equal volume of 4% SDS and 4% Triton X100). The bottle with its content was agitated for a contact time of 24 h in a Heldoph rotary shaker.<sup>19</sup> The residues obtained after centrifugation for 10 min at 10000 rpm were air-dried at room temperature.<sup>26</sup> Each of these was pulverized and kept in a desiccator prior to heavy metal, radionuclide and microbe determination. This washing removes arsenic compounds, pesticides and other contaminants.<sup>26</sup>

# Determination of heavy metals, radionuclides and microbes after remediation

After the remediation processes, the organoleptic properties, levels of heavy metals, radionuclides and microbial load of the clay samples were assessed following the procedures undertaken before the remediation; in order to appraise the efficiency of removing each of the contaminants studied.

#### Statistical analysis

Paired t-test and ANOVA were used to statistically compare the results obtained for the organoleptic properties, level of heavy metals, radionuclides and microbes in the clay before and after remediation with significance taken at P < 0.05.

## **Results and Discussion**

Organoleptic properties of non - remediated samples and those remediated with different solutions

## Effect on pH of the clay

The pH of edible kaolin obtained from Katsina State, Nigeria (EKK) was 7.29, compared to pH 6.5 for the one purchased (EKM) from Samaru and Sabo markets in Zaria, Nigeria (Figure 1).

Remediation resulted to increased pH of the clay, regardless of the solution used for the remediation, except for the use of HCl where the

pH values were 4.06 and 5.38 in edible kaolin obtained from Kankara, Nigeria – EKK and edible kaolin purchased from market – EKM respectively. Notwithstanding, the change in pH of the clay samples due to all the remediating solutions is still within the values of pH reported for edible earth material by Costarramone *et al.*<sup>27</sup> being 4.9, and pH 8.91 limit by Kim *et al.*<sup>28</sup>.From the study, the use of water, CaCl<sub>2</sub> and surfactant solutions altered the pH values to become alkaline (Figure 1). This is because water has a pH of 7, CaCl<sub>2</sub> has Ca<sup>2+</sup>,<sup>29</sup> also the surfactant solution is basic containing sulphate and hydroxyl (OH<sup>-</sup>) ions.

## Effect on colour of the clay

The clay samples – EKK and EKM – recorded a colour of 15 units on Hezan scale of the lovibond 1000 colour comparator. Also, colour of the clay samples did not change with remediation, except the sample remediated with surfactant solution, which had the colour being more intense (70 units) (Figure 2).

#### Effect on taste of the clay

The result of the panellists indicated that the purchased edible kaolin sample (EKM) was moderately sour (Figure 3). Remediation resulted to change of the taste of the clay. The surfactant solution and water made it tasteless after remediation, whereas HCl and CaCl<sub>2</sub> did not change the taste of the clay sample (Figure 3).

## Remediation of heavy metals

The study indicated that the concentration of As in EKM was higher being  $1.31 \pm 0.05$  mg/kg. The best solution for the extraction of As from EKM was 0.1M CaCl<sub>2</sub>, this extracted 0.28 mg/kg As which is 21% of the accumulated As. This was followed by HCl which reduced the concentration of As from  $1.31 \pm 0.05$  mg/kg in EKM to  $1.07 \pm 0.07$ mg/kg, indicating 18% extraction. This was closely followed by the surfactant solution that extracted 7% As (0.09 mg/kg As from EKM). Water was the least extractant and it extracted 0.02 mg/kg As (2%).

Therefore, remediation of As from the edible kaolin was in the order:  $CaCl_2(21\%) > HCl(18\%) > SDS/T(7\%) > H_2O(2\%)$  (Figure 4).

On the other hand, HCl was best for the extraction of As from EKK (Figure 4), this extracted 0.23 mg/kg (19%) As. This was followed by water which extracted 0.13 mg/kg (10.7%) of As. The surfactant solution was next and it reduced the concentration of As from 1.21  $\pm$  0.01 mg/kg to  $1.18 \pm 0.07$  mg/kg in EKK, thereby extracting 0.03 mg/kg (2%) of As. While CaCl<sub>2</sub> was the least extractant of As from EKK, this extracted 0.02 mg/kg (2%) As. So the remediation process for As from kaolin from Kankara was in the order: HCl (19%) > H<sub>2</sub>O> SDS/T100 (2%)> CaCl<sub>2</sub> (2%). So, local processing of edible clay should use acid as the extractant.

Therefore, CaCl<sub>2</sub> was the most efficient solution for the removal of As in EKM and HCl was the best for EKK. However, the concentration of As in the raw and remediated clay samples was not statistically significant (P > 0.05).

The concentration of Cr in EKK was  $1.184 \pm 0.22$  mg/kg compared to EKM which was  $0.814 \pm 0.009$  mg/kg. For EKK water was best for extraction of Cr, this extracted 1.156 mg/kg Cr being98% of the Cr. HCl was next, this reduced the concentration of Cr from  $1.184 \pm 0.22$  mg/kg to  $0.0.032 \pm 0.002$  mg/kg, being 97 % of Cr. This was followed by CaCl<sub>2</sub> and surfactant solution that both extracted 1.15 mg/kg (97 %) of Cr from EKK (Figure 5).

The trend for remediation of Cr from edible kaolin from Kankara was in the order: water  $(98\%) > HCl (97\%) > CaCl_2 = SDS/T (97\%)$ . The best extractant for chromium from edible kaolin is water.

Water was also the best for the extraction of Cr from EKM, this extracted 0.787 mg/kg (97 %) of Cr. This was followed by HCl which reduced the concentration of Cr from  $0.814 \pm 0.009$  mg/kg to  $0.031 \pm 0.001$  mg/kg, thereby extracting 0.783 mg/kg (96%) of Cr. This is followed by the use of surfactant solution, which extracted 0.775 mg/kg (95%) of Cr. While the least extraction of Cr from EKM was by CaCl<sub>2</sub>, this extracted 0.751 mg/kg (92%).

The trend for Cr removal from EKM was in the order:  $H_2O$  (97%) > HCl (96%) > SDS/T100 (95%) >  $H_2O$  (95%). Hence water was the best extractant of Cr in EKM.

For the remediation of Cr from EKK, water was the most efficient extractant, it removed 98% of the total Cr concentration of 2.368 mg/kg. This was followed by HCl which extracted 0.783 mg/kg (97%) of Cr. However, it has been reported that strong acids attack and degrade the soil crystalline structure at extended contact times.<sup>22</sup>The surfactant

solution extracted 0.775 mg/kg (95%) and the least was  $CaCl_2$  which extracted 0.751 mg/kg (92%) of Cr. Therefore, as depicted in Figure 5, the remediation of Cr in edible kaolin fom Farar kasa hill, Kankara was in the order:  $H_2O > HCl > SDS/T100 > CaCl_2$ .

The study indicated that  $H_2O$ , HCl and CaCl<sub>2</sub> resulted to significant reduction in the level of Cr in the clay sample.

The level of Pb in EKK was  $0.167 \pm 0.008$  mg/kg and was  $0.288 \pm 0.021$  mg/kg in EKM (Figure 6). This could have resulted from the geological composition of the clay.

For EKK, water was the best extractant of Pb as it extracted 0.155 mg/kg (93%). HCl was the next, this extracted 0.125 mg/kg (75%) of Pb. While CaCl<sub>2</sub>reduced the Pb content from 0.167 mg/kg to 0.043 mg/kg, indicating 74% reduction. The least extractant was the surfactant solution which extracted 0.094 mg/kg (56%) of Pb. So, water was the best for the remediation of Pb from EKK.

As depicted in Figure 6, HCl was best for the extraction of Pb from EKM, this extracted 0.125 mg/kg (70%) of Pb. This was followed by water which reduced the concentration of Pb from 0.288  $\pm$  0.021 mg/kg to 0.095  $\pm$  0.007 mg/kg in EKM, thereby extracting 0.193 mg/kg (67%) of Pb. Followed by this was CaCl<sub>2</sub> which extracted 0.188 mg/kg (65%) of Pb from EKM. CaCl<sub>2</sub> resulted to the least remediation of Pb from EKM. this removed 61% of the Pb. The study showed that the remediation of Pb from EKM follows the order: HCl (70%) > SDS/T100 (67%) > H<sub>2</sub>O (65%) > CaCl<sub>2</sub> (61%). Hence HCl was the best extractant of Pb from EKM.

This report supports the study of Reddy and Chinthamreddy,<sup>29</sup> in which changes in Ni, Cu, Zn, Cd, and Pb speciation and uptake by maize in a sandy loam has EDTA and citric acid as the potential chelating agents for remediating the soil.<sup>19</sup> And also supports the work of Khodadoust *et al.*<sup>30</sup> in the evaluation of different extracting solutions for the removal of phenanthrene, lead and zinc from a contaminated soil. Chelating agents (ethylenediamine tetra acetic acid, EDTA and diethylene triamine pentaacetic acid, DTPA) and selected acids effectively extract Pb and Zn.<sup>27</sup> In addition, the lead being extracted could have existed as a soluble compound of lead.

The study showed that there was significant reduction in the Pb contents of the two clay samples by using HCl (P < 0.05).

#### Remediation of Radionuclides

The result of the remediation of <sup>232</sup>Th from EKM (Figure 8) indicates that the activity concentration of <sup>232</sup>Th in the raw clay samples was 43.57 ± 4.21 Bq/kg. Remediation with the surfactant solution was best for the remediation of <sup>232</sup>Th, this led to the removal of 22.13 Bq/kg (46%) of <sup>232</sup>Th. This was followed by HCl which reduced <sup>232</sup>Th from 43.57±4.21 to 39.02±4.67 Bq/kg (10%) of <sup>232</sup>Th. On the other hand, H<sub>2</sub>O extracted 0.8 Bq/kg (2%), while CaCl<sub>2</sub> reduced the activity concentration of <sup>232</sup>Th to 43.01±5.03 Bq/kg being 1% reduction. From the study, remediation of <sup>232</sup>Th from edible kaolin (EKM) followed the order: SDS/T100(46%) > HCl (10%) > H<sub>2</sub>O (2%) > CaCl<sub>2</sub> (1%).

The result of the remediation of  $^{228}$ Ra from the Edible Kaolin purchased from Samaru market, Zaria, Nigeria is presented in Figure 7. The activity concentration of  $^{228}$ Ra in the raw clay samples was  $53.03 \pm 8.66$  Bq/kg. After remediation, CaCl<sub>2</sub> was best for the remediation of  $^{228}$ Ra as it extracted 8.87 Bq/kg (17%) of  $^{228}$ Ra.

This was followed by HCl which reduced the activity concentration of  $^{228}$ Ra from 52.03±8.66 to 43.53±7.88 Bq/kg thereby extracting 8.5 Bq/kg (16%) of  $^{228}$ Ra. On the other hand, the use of surfactant solution extracted 4.2 Bq/kg (8%) Ra, while water led to the extraction of 2.41 Bq/kg (4.6%)  $^{228}$ Ra. So,  $^{228}$ Ra was best extracted from edible kaolin by CaCl<sub>2</sub> and HCl.

Students' t-test showed that treatment does not significantly reduce the activity concentration of <sup>228</sup>Ra in the clay samples.

The activity concentration of  $^{238}$ U in the raw edible kaolin samples was 25.18  $\pm$  2.61 Bq/kg. Remediation with the use of surfactant solution resulted to removal of 8.88 Bq/kg (35%) of  $^{238}$ U. This was followed by water, which reduced the activity concentration of  $^{238}$ U from 25.18  $\pm$  2.61to 19.11  $\pm$  2.08 Bq/kg, thereby extracting 24%  $^{238}$ U. On the other hand, HCl extracted 4.64 Bq/kg (18%), while CaCl<sub>2</sub> extracted 2.06 Bq/kg (5%) of  $^{238}$ U. Therefore, removal of  $^{238}$ U from 'Nzu' clay follows the ranking: SDS/T100 (35%) > H<sub>2</sub>O (24%)> HCl (18%) > CaCl<sub>2</sub> (5%). With the best extractant for  $^{232}$ Th from EKM clay being the surfactant solution.

There was no significant reduction in the level of  $^{238}$ U in the clay after remediation (P > 0.05).

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**Figure 1:** pH of the edible kaolin from Kankara with remediation. EKK = Edible Kaolin Kankara, EKM = Edible Kaolin Market, NR = Non – remediated.

![](_page_3_Figure_4.jpeg)

Figure 2: Colour of edible kaolin with remediation.

![](_page_3_Figure_6.jpeg)

Figure 3: Taste of the clay samples with remediation.

Key: For the taste (x) axis, 1 = sour, 2 = moderately sour, 3 = tasteless, 4 = moderately sweet, 5 = sweet.

![](_page_3_Figure_9.jpeg)

Figure 4: Remediation of As from edible kaolin.

![](_page_3_Figure_11.jpeg)

Figure 5: Remediation of Cr from kaolin.

![](_page_3_Figure_13.jpeg)

Figure 6: Remediation of Pb from kaolin sample.

![](_page_4_Figure_2.jpeg)

Figure 7: Remediation of <sup>232</sup>Th in Edible Kaolin.

![](_page_4_Figure_4.jpeg)

Figure 8: Remediation of <sup>228</sup>Ra from Edible Kaolin.

![](_page_4_Figure_6.jpeg)

Figure 9: Remediation of <sup>238</sup>U from edible Kaolin.

![](_page_4_Figure_8.jpeg)

Figure 10: Microbial load of the edible kaolin samples.

Bacterial count of non - remediated samples and those remediated with different solutions

From Figure 10, all the clay samples were contaminated by bacteria. The range of contamination of the raw clay by bacteria was from  $4 \times 10^5$  to  $7.6 \times 10^5$  CFU/mL.

For the edible Kaolin obtained from Kankara in Katsina State – Nigeria (EKK), CaCl<sub>2</sub> was the best extractant as it reduced the bacteria load to  $1.1 \times 10^5$  CFU/mL (72%) of the bacteria. This was followed by HCl which reduced the bacteria load from  $4.0 \times 10^5$  CFU/mL to  $1.5 \times 10^5$  CFU/mL, while the surfactant solution reduced it to  $1.8 \times 10^5$  CFU/mL (55%) of the bacteria load, water led to the reduction by  $0.5 \times 10^5$  CFU/mL (13%).

Therefore, the treatment is also effective in reducing the bacterial contaminants of the edible clay.

The edible kaolin purchased from Samaru market, Zaria (EKM), had CaCl<sub>2</sub> as the best extractant, this reduced  $6.3 \times 10^5$  CFU/mL (83%) of the bacteria. This was followed by HCl which reduced the bacteria load from  $7.6 \times 10^5$  CFU/mL to  $2.5 \times 10^5$  CFU/mL being 67%. Water followed with extraction of  $5 \times 10^5$  CFU/mL (66%) of the bacteria load. And the least extractant was the surfactant solution which extracted  $4.2 \times 10^5$  CFU/mL (55%).

Therefore, the removal of bacteria from EKM was in the order:  $CaCl_2$  (83%) > HCl (67%) > H<sub>2</sub>O (66%) > SDS/T100 (55%). Hence,  $CaCl_2$  was best for remediating the edible clay, since it gave the highest percentage reduction of bacteria. This is very relevant for health implication of ingesting the edible clay.

## Conclusion

From the study, the pH of the edible clay samples increased when remediated with the surfactant solution,  $CaCl_2$  and water. The taste of the clay was also slightly affected after remediation. The clay samples contained As  $(1.31 \pm 0.05 \text{ to } 1.21 \pm 0.05 \text{ mg/kg})$ , Cr  $(1.184 \pm 0.22 \text{ to } 0.814 \pm 0.009 \text{ mg/kg})$ , and Pb  $(0.288 \pm 0.021 \text{ to } 0.167 \pm 0.008 \text{ mg/kg})$ . Remediation with 0.1M HCl was the best for removal of As; water was the best for extraction of Cr and Pb. Also  $0.1M \text{ CaCl}_2$  was the best for removing  $^{232}$ Th and  $^{238}$ U. All the clay samples were contaminated with bacteria; and remediation resulted to removal by 79%. Therefore, the study proffers a remediating scheme for edible kaolin clay, in order to maintain the medical benefits and minimize ingestion of contaminants by addicted consumers.

## **Conflict of interest**

The authors declare no conflict of interest.

## **Authors' Declaration**

The authors hereby declare that the work presented in this article is original and that any liability for claims relating to the content of this article will be borne by them.

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