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Heavy Metals Solubilization in EDTA-Assisted Remediation of Sewage Sludge

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ABSTRACT

The effect of various EDTA masses on metal removal from sludge when digested with HNO_3 and H_2SO_4 acids at various pH levels was studied. Results of mean concentration (mg/l) of Cd, Mn, Pb, Ni, and Cu for acid digestion of sewage sludge using HNO_3 and H_2SO_4 at pH 2.0, 3.0, and 5.0 at various EDTA masses (1.0, 1.5 and 2.0 g) showed that Cd was the most extracted metal (1.0–19%) at pH 2.0. At pH 3.0 and 5.0 for the same EDTA weights, the range of Cd was 3.0–21% and 4.0–30% using HNO_3 and H_2SO_4 acids as extractant, respectively. There was a slight increase in the amount of Cd extracted at pH 5.0 using H_2SO_4 for digestion. When HNO_3 was used for digestion, there was an increase in the amount of Cd removed from the sludge as the amount of EDTA was increased from 1.0 to 1.5 to 2.0 g at all pH levels. This trend was not replicated using H_2SO_4. Pb and Cu were poorly solubilized at all pH levels for all EDTA masses using both acids. Nevertheless, Cu was the least extracted metal overall, totaling 0.0312, 0.0335, and 0.0367 mg/l at 1.0, 1.5, and 2.0 g EDTA masses at pH 3.0 as opposed to 0.0375 mg/l Cu extracted in the control experiment at the same pH level using HNO_3.

Keywords: chelate, complexing, contamination, digestion, extraction, mobilization

INTRODUCTION

Heavy metal (HM) solubilization efficiency is determined by the appropriate application of inorganic and organic acids. These conditions include pH, redox potential, and reaction time. To promote solubilization of HM, it is necessary to decrease the pH of the sludge to about 1.0–2.0 (Ukiwe and Oguzie 2008). At this pH, formation of soluble metal complexes and oxidizing insoluble reduced metal forms to soluble forms are favored. Chelates such as EDTA (ethylenediamine tetracetic acid) and NTA (nitrilotriacetic acid) are used to enhance metal solubilization. It has been demonstrated that about 50% Cr, Pb and Zn were extracted by EDTA at a pH value of 4.5 and 20°C (Van and Vandecasteele 2001). However, this was not the best extracting condition for Zn, which was better released by a reducing agent at pH 2.0 and 96°C. Moreover, EDTA did also not satisfactorily remove Cu. The efficiency of chelating agents EDDS (ethylenediamine disuccinic trisodium salt) and EDTA have been investigated in solubilization of HM from soils contaminated by the addition of organic and inorganic residues. Results showed that metals were highly available in the soil fractions when suspended in chelating agents for 24 hrs. The metal solubilized varied according to soil type, the chelating agent added and the specific metal. This study claimed that EDDS solubilized more Cu than EDTA in soil samples.

In the past few years, EDDS has received some attention due to its greater rate of degradation and its strong chelating characteristics comparable to EDTA, although its biodegrading rate varies largely depending upon the examined conditions (Evangelou *et al*. 2007). EDDS has also been reported to be more effective than EDTA in extracting metals most specifically in removal of Ca in sludge (Hauser *et al*. 2005; Luo *et al*. 2005). The fractionation of HM in sludge is a technique extensively used to enable a better understanding in metal binding to the sludge solid phase, which ultimately reflects its availability. Usually, there is a general agreement that the metals found in the soluble/exchangeable phase and in the organic matter fraction are more readily available for mobilization and consequently, more susceptible to binding to chelating agents. The addition of metals to soil due to the repetitive application of sewage sludge leads to accumulation where the metals remain in the most labile fractions of the soil, mainly due to organic matter in the system (Galdos *et al*. 2004). Assisting chelating agents such as EDTA may be limited to clean up Pb from soil leachate due to its low solubility in the phytoextraction process (Coscione *et al*. 2009). Pereira *et al*. (2007) evaluated the effect of EDTA on maize grown in soil containing Pb and observed that EDTA did not effectively solubilize Pb since maize grown on EDTA-amended soil was shown to have a 52% uptake of Pb while that grown on soil without EDTA had 57% Pb uptake. The study further noted that the poor uptake of Pb was due to its low solubilization potential. Several factors should be considered when comparing chelating agents for HM decontamination (Tandy *et al*. 2004). These factors include, among others; the ratio of chelating agent/HM. It was noted that a high concentration of chelating agents can alter soil nutrient availability causing HMs to leach. However, using smaller quantities of chelating agents generates competition of chelates with other complex substances found in soil which are sometimes in much larger concentrations than the metal contaminants. EDTA has been demonstrated to show high removal efficiencies for Fe, Ni and Cd at neutral pH (Samanidou and Fytianos 1990). The addition of EDTA to sludge based on stoichiometric requirements for metals such that the percentage of metal removal by EDTA does not correspond to stability constants for the EDTA complexes raises issues associated with HM bound by various ligands and exist as various insoluble salts (Samanidou and Fytianos 1990). HMs can be mobilized from sludge particles by changes in pH and oxidation-reduction potential conditions and by complexing with synthetic complexing agents (NTA and EDTA), inorganic complexing agents, or with natural

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Research Note

chelating agents such as soluble humic substances (Samanidou and Fytianos 1990). The distribution of metals in sludge depends on the chemical properties of the specific metal and on the sludge characteristics which are determined by the presence of complexing agents such as EDTA. Interaction experiments (Peñalosa *et al.* 2007) have indicated that EDTA and NTA are more efficient than malate and citrate in solubilizing metals (Fe, Mn, Cu, Zn, Cd), with minimum differences between EDTA and NTA. A pot trial showed that NTA was able to mobilize toxic elements (Mn, Cu, Zn, As, Cd) from sludge-polluted soil, and hence increasing their concentrations in plants. However, the NTA treatment promoted an increase of toxic element concentrations, especially for As, Cd, and Pb exceeding the maximum permissible levels. So a careful management of chelate is necessary when used (Peñalosa *et al.* 2007).

pH is one of the most powerful parameters to control the transfer of HMs from immobile solid phase forms to more mobile, and therefore, more bioavailable solution phase forms. This parameter influences adsorption equilibria, the stability of organomineral complexes and oxidation-reduction potential. Veeken and Hamelers (1999), and Sims and Klime (1991) noted that about 57% of Cr was extracted by EDTA at a pH value of 4.5. Consequently, when complexing agents such as citric acid, oxalic acid or EDTA are applied to sludge, Cr can be solubilized at a higher pH value (3–5) than when a strong acid such as HNO_3 was applied. However, pot and leaching column experiments conducted to optimize chelator-assisted extraction of Pb from contaminated soils revealed that optimum extraction occurred at added EDTA concentration of 5 mM per dose in low Pb soils for 10 days, while for high Pb soil, 10 mM of EDTA was added for five intermittent doses for 7 days. Concluding, it was noted that combined with column experiments, chelator-assisted extraction is more suitable for slightly metal contaminated soils (Liu *et al.* 2008).

The main objective of the present study was to assess the effect of various EDTA weights on metal removal from sludge when digested with HNO_3 and H_2SO_4 acids at various pH levels.

MATERIALS AND METHODS

The sewage samples used in this experiment were obtained from a septic tank of the male ward of the Federal Medical Center, Owerri, Nigeria. The tank has a capacity of about 800 m^3/day. A 50-L plastic container previously washed and rinsed with deionised water was used to collect the sewage sample from the tank. The content was then transported to the laboratory of the Department of Chemistry of the Federal University of Technology, Owerri, Nigeria and the container was allowed to stand for 24 hrs for solid particles to settle. From this stock solution samples were continually drawn. A quantity of sample containing 2 L of sewage was measured into a 2 L beaker previously washed and rinsed with deionised water. From this 2 L sewage sample about 500 ml of sewage was measured and poured into a 1000 ml beaker and centrifuge (Micro Centrifuge Model 5415C) for 2 hrs at 300 rpm at 28°C. The resulting solution was filtered through Whatman No. 42 filter paper. To 30 ml of the filtrate 10 ml of a 50% (v/v) HNO_3 solution was added and the mixture was stirred continuously for 5 min. The pH of this mixture was maintained at 2.0 with the addition of some amount of 10 M NaOH. About 1.0 g of EDTA was further added to the mixture and stirred for another 5 min. The mixture was then heated at 80°C for 1 hr with intermittent addition of deionised water to prevent drying up of the mixture. The mixture was further cooled, filtered through Whatman No. 42 filter papers. The resulting solution was made up to 100 ml with deionised water in a 100 ml standard flask and 5 ml of this solution was used for analysis by atomic absorption spectrophotometry (AAS) using a Model SOLAAR V10 atomic absorption spectrophotometer for the elements Cd, Mn, Pb, Ni and Cu. Three repetitions were made and the mean concentration (mg/l) of HM was obtained by the method described by Ukiwe and Oguzie (2008). This procedure was repeated (control experiment) without the addition of EDTA. However, the above procedure was repeated for 50% (v/v) HNO_3 acid and at 1.0 g EDTA and without EDTA (control experiment) for pH 3.0 and 5.0. Also, at 50% (v/v) HNO_3 acid concentration, the procedure was repeated for 1.5 and 2.0 g EDTA and without EDTA (control experiment) at pH 2.0, 3.0, and 5.0. The entire procedure above was repeated at 1.0, 1.5, and 2.0 g EDTA and without EDTA content (control experiment) at pH 2.0, 3.0, and 5.0 using 50% (v/v) H_2SO_4.

Statistical analysis

Data are presented as arithmetic mean, standard deviation and standard error of the mean. The standard error of the difference between mean pH with EDTA and without EDTA (control experiment) was employed to measure difference at various pH levels. The generalized *t*-test was also used to estimate if there was significance difference between the amounts of HMs solubilized with and without EDTA at the various pH levels.

RESULTS AND DISCUSSION

EDTA is an aminopolycarboxylic acid containing six donor atoms, which acts as hexadentate ligands. This compound forms strong and very stable complexes with many metal cations (divalent and trivalent ions), increasing the metal ion solubility in aqueous solution as water-soluble complexes (Baeze *et al.* 2007). EDTA use has become widespread recently due to its cheap value and most suitable for multipurpose applications in moderating the adverse effects of transition metal ions that result due to the activity of detergents, cosmetics and photochemicals. EDTA is used in textile manufacture, hydrogen peroxide, and ozone based cellulose bleaching processes and in the treatment of soils contaminated with HM (Ekland *et al.* 2002). EDTA's poor biodegradation has been a limiting factor to its use worldwide (Pitter and Sŷkora 2001). Chemical pretreatment to degrade EDTA into biodegradable species, represents an interesting choice to improve the efficiency of biological effluent systems (Oviedo *et al.* 2003).

Tables 1-6 give mean concentrations (mg/l) of Cd, Mn, Pb, Ni, and Cu for acid digestion of sewage sludge using HNO_3 and H_2SO_4 at pH 2.0, 3.0, and 5.0 at various EDTA masses of 1.0, 1.5, and 2.0 g respectively. Cd was the most extracted metal totaling 0.5 – 19% for the above EDTA masses respectively at pH 2.0 using HNO_3 and H_2SO_4 acids for digestion. At pH 3.0 and 5.0 for the same EDTA masses, Cd extraction ranged 1.2 – 21% and 4.0 – 30% for HNO_3 and H_2SO_4 respectively, using the above acids in the same order. There was a slight increase in the amount of Cd extracted at pH 5.0 using H_2SO_4 for digestion. It was also noticed that using HNO_3 for digestion, there was an increase in trend in the amount of Cd removed from the sludge as the amount of EDTA was increased from 1.0, 1.5, to 2.0 g at all pH levels. Though, this trend was not replicated using H_2SO_4. Pb and Cu were poorly solubilized at all pH levels for all EDTA concentration using both acids. Nevertheless, Cu was the least extracted metal overall and the least extracted metal overall at pH 3.0 totaling (0.0312, 0.0335, 0.0367 mg/l) at 1.0, 1.5, and 2.0 g EDTA masses respectively as opposed to 0.0375 mg/l Cu extracted in the control experiment at pH 3.0 using HNO_3.

Tables 1-6 also present data of the standard deviation and standard error of mean concentration (mg/l) of HMs at various HM weights at different pH levels using HNO_3 and H_2SO_4, respectively. The generalized *t*-test between set observations (HNO_3 and H_2SO_4 at various HM weights at different pH levels) and control experiment are also given. These values were tested separately at 8 degrees of freedom, $P < 0.01$. It can be deduced from the facts above that the presence of EDTA did not significantly aid in extracting HMs when compared to ordinary extraction using acids only. The reason why Pb and Cu were poorly extracted from the sludge in the present study could be ascribed to some factors which include the fact that these two metals are always tightly bound to the organic and inorganic matter fraction in sludge resulting in very small proportion being

Table 1 Concentration of heavy metals in sewage at pH 2.0 using HNO_3.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0529 ± 1.7	9.8	0.0560 ± 1.1	6.3	0.0565 ± 2.0	0.1	0.0456 ± 4.0	0.2
Mn	3.4650 ± 0.8	4.6	6.8001 ± 0.3	1.7	2.9734 ± 0.6	3.4	5.5037 ± 1.8	0.1
Pd	0.2449 ± 0.8	4.6	0.1075 ± 1.0	5.7	0.1313 ± 0.9	5.2	0.4160 ± 1.1	6.3
Ni	0.1829 ± 7.4	0.4	0.1657 ± 1.8	0.1	0.1852 ± 1.9	0.1	0.1694 ± 5.2	0.3
Cu	1.9458 ± 0.9	5.2	1.6553 ± 4.0	0.2	0.1791 ± 1.0	5.7	0.1764 ± 2.7	0.1

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.06; (b) and (z) = 0.29; (c) and (z) = 0.45

Table 2 Concentration of heavy metals in sewage at pH 3.0 using HNO_3.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0517 ± 9.3	0.5	0.0535 ± 1.6	9.2	0.0534 ± 2.0	0.1	0.0425 ± 1.5	8.6
Mn	2.9310 ± 0.8	4.6	1.1401 ± 1.3	7.5	0.7702 ± 7.2	0.4	0.6816 ± 1.0	5.7
Pd	0.0500 ± 1.1	6.3	0.0644 ± 0.7	4.0	0.1462 ± 0.8	4.6	0.2433 ± 1.9	0.1
Ni	0.2714 ± 1.9	0.1	0.2288 ± 4.5	0.2	0.2855 ± 1.0	5.7	0.1742 ± 6.7	0.3
Cu	0.0312 ± 4.2	0.2	0.0335 ± 2.0	0.1	0.0367 ± 5.7	0.3	0.0375 ± 3.6	0.2

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.73; (b) and (z) = 0.28; (c) and (z) = 0.13

Table 3 Concentration of heavy metals in sewage at pH 5.0 using HNO_3.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0476 ± 1.3	7.5	0.0488 ± 1.0	5.7	0.0497 ± 1.0	5.7	0.0411 ± 9.2	0.5
Mn	1.5249 ± 1.0	5.7	1.1505 ± 0.7	4.0	0.6100 ± 0.5	2.8	1.1817 ± 1.1	6.3
Pd	0.2129 ± 1.0	5.7	0.2862 ± 0.9	5.2	0.1959 ± 1.2	6.9	0.4945 ± 1.8	0.1
Ni	0.2534 ± 0.6	3.5	0.2013 ± 3.0	0.1	0.2877 ± 1.6	9.2	0.1697 ± 5.5	0.3
Cu	0.1824 ± 1.0	5.7	0.1006 ± 1.0	5.7	0.1429 ± 9.4	0.5	0.1879 ± 1.5	8.6

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.08; (b) and (z) = 0.19; (c) and (z) = 0.71

Table 4 Concentration of heavy metals in sewage at pH 2.0 using H_2SO_4.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0598 ± 5.6	0.3	0.0532 ± 1.0	5.7	0.0575 ± 1.0	5.7	0.0529 ± 4.1	0.2
Mn	1.5478 ± 2.2	0.1	1.7796 ± 2.7	0.1	3.0590 ± 0.4	2.3	1.5469 ± 3.6	0.2
Pd	0.3132 ± 1.1	6.3	0.3110 ± 1.9	0.1	0.3035 ± 1.8	0.1	0.3532 ± 1.0	5.7
Ni	0.1769 ± 4.0	0.1	0.2189 ± 2.4	0.1	0.1951 ± 2.2	0.1	0.0124 ± 3.0	0.1
Cu	0.4855 ± 1.9	0.1	0.4971 ± 1.6	5.7	0.1168 ± 1.0	5.7	0.0473 ± 1.2	6.9

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.28; (b) and (z) = 0.39; (c) and (z) = 0.52

Table 5 Concentration of heavy metals in sewage at pH 3.0 using H_2SO_4.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0482 ± 1.9	0.1	0.0491 ± 4.4	0.2	0.0552 ± 1.8	0.1	0.0476 ± 7.3	0.4
Mn	0.2073 ± 1.0	5.7	0.2096 ± 8.0	0.4	0.2404 ± 6.6	0.3	1.1426 ± 2.4	0.1
Pd	0.2834 ± 1.4	8.0	0.2826 ± 2.0	0.1	0.2431 ± 1.1	6.3	0.2722 ± 2.0	0.1
Ni	0.1783 ± 2.3	0.1	0.2036 ± 2.1	0.1	0.1767 ± 1.8	0.1	0.0360 ± 2.6	0.1
Cu	0.0802 ± 4.2	0.2	0.0754 ± 1.9	0.1	0.0384 ± 1.4	8.0	0.0932 ± 5.7	0.3

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.73; (b) and (z) = 0.70; (c) and (z) = 0.76

Table 6 Concentration of heavy metals in sewage at pH 5.0 using H_2SO_4.

EDTA content	**1.0 g$^{(a)}$**		**1.5 g$^{(b)}$**		**2.0 g$^{(c)}$**		**Control experiment (without EDTA)$^{(z)}$**	
Heavy metals	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**	**Mean (mg/l) ± SD × 10^{-1}**	**SEM×10^{-2}**
Cd	0.0476 ± 2.1	0.1	0.0575 ± 1.4	8.0	0.0530 ± 9.7	0.5	0.0397 ± 4.2	0.2
Mn	1.2754 ± 4.7	0.2	3.6447 ± 0.1	0.0	0.2355 ± 4.6	0.2	0.7649 ± 0.6	3.4
Pd	0.2094 ± 0.9	5.2	0.2092 ± 1.2	6.9	0.2859 ± 1.5	8.6	0.2883 ± 1.2	6.9
Ni	0.1281 ± 1.0	5.7	0.1296 ± 1.0	5.7	0.1626 ± 6.8	0.3	0.0349 ± 2.9	0.1
Cu	0.0820 ± 5.2	0.3	0.0754 ± 3.4	0.2	0.0544 ± 2.2	0.1	0.1107 ± 3.7	0.2

SEM: Standard Error of the Mean; *t*-test between (a) and (z) = 0.36; (b) and (z) = 0.78; (c) and (z) = 0.60

available to be solubilized and also because of the low pH adopted in the present study. It has being reported that Pb and Cu have been successfully extracted at pH above 5.5 (Van and Vandecasteele 2001). A chemical washing process applied to dredge sea and river sediments to investigate the presence of HM has shown that for a given experimental condition, the remediation efficiency was strongly dependent on the specific contaminant under consideration, with contaminant speciation and distribution in the solid matrix as well as affinity for the extracting agent playing a major role in the decontamination process using five chelating agents which included EDTA (Polettini *et al.* 2008). The study went on to argue that in the presence of multiple contaminants, the use of a single extraction agent is not sufficient to attain adequate quality levels for the treated sediments. To solve this problem, advanced oxidation processes for remediation of HM and radionuclides contaminated soils and sediments in a closed process have been proposed (EPA 2009). Chelating agents in a washing solution are used to wash metals and radionuclides from the soil/sediments as chelate complexes. Supporting compounds in the washing solution remove organic pollutants. Advance oxidation, preferably, ozonation or ultra violet radiation is used for degradation of complexes and organic pollutants in the washing solution. Released metals/radionuclides are absorbed and the treated washing solution is reused for soil/sediment rinsing in a closed process loop, to remove residual complexes, and as a medium for the chelating agent and supporting compounds in multiple washing steps. Remediated soil/sediment is returned to the original place or deposited. The process saves water, produces no harmful emissions and is gentle to the soil. The use of plants in phytoremediation studies in EDTA assisted HM solubilization of sewage sludge have achieved much success and have been a promising alternative to the use of EDTA in HM recovery from sludge. A study investigating the effects of addition of chelates to soil showed that when chelating agents were added to soil to solubilize the metals for enhanced phytoextraction, the mobility of HM in biosolids in the soil placed in column that have plants and columns without plants in a greenhouse from a sludge farm was affected. With or without plant, the EDTA mobilized all HM (Cd, Cu, Zn) and increased their concentration in drainage water when drainage water was analyzed for the three toxic HM after plant seedlings grown for 144 days was irrigated onto the surface of the soil at a rate of 1 g/kg of soil with EDTA (Liphadzi and Kirkham 2006).

Studies on the use of EDTA assisted HM removal from sewage sludge is rare and the present study would advise on more studies in this subject.

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