

Removal of cesium from contaminated soils using EDTA-Na and DTPA chelating agents

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Abstract

Soil contamination with Cs possesses a serious environmental threat; and therefore, its remediation is an obligation to ensure environmental safety. The current study aims at evaluating the efficiency of using either EDTA-Na or DTPA as chelating agents for removal of Cs from contaminated soils. Three soils (a clayey one and two sandy loam soils differing in the organic matter and CaCO₃ contents) were selected to attain this aim. Samples of the soils under study were artificially contaminated with CsCl at three levels i.e. 50, 100, and 200 mg Cs kg⁻¹. These samples were subjected to sequential extraction pre- and post-leaching to find out Cs redistribution among the different soil fractions after soil washing. The implications of soil washing with the investigated chelating agents on decreasing the risk assessment hazards associated with Cs hazards are discussed in this study. Key results indicated that DTPA was more efficient in removal of Cs from the contaminated soils than the EDTA-Na. Both decreased considerably soil Cs, particularly from those which are considered the mobile fractions (water soluble and exchangeable fractions) beside of the carbonate bound fractions. Accordingly, the calculated risk assessment code declined noticeably from “very high risk” levels (more than 50) to “medium risk” levels (ranging from 30 to 50) for most spiked soils. In conclusion, DTPA and EDTA-Na are effective chelating agents in reducing the risks associated with Cs contamination in soils.

Keywords: Cesium fractionation, washing, risk assessment code, EDTA-Na, DTPA.

1. Introduction

Accumulation of potentially toxic elements (PTEs) in soil represents a serious environmental threat worldwide [1-7]. Cesium is one of the main fission products, which has been started to release to the atmosphere after the first nuclear bomb at the sixties of the last century [8-10]. The following nuclear disasters such as Chernobyl in 1986 and Fukushima in 2011 returns the fear of the negative consequences of Cs on the environment [11-13]. Cs as a radioactive nuclide (¹³⁷Cs has about 30 years half-life) risks are higher than the corresponding ones of the stable toxic form [14]. Generally, the chemical behavior of ¹³⁷Cs is similar to those of Na and K [15]; thus it deposits in the soft tissues of aquatic and terrestrial organisms [16]. In the human body, it harms the central nervous system and causes kidneys and liver failure [17].

Sorption of cesium on soil particles and its transfer to plant are affected by various factors [18-19], such as soil pH which diminishes Cs mobility at high pH values [20]. Also, concentration of potassium lessens Cs sorption on soil particles due to collapse of the hydrated and expanded interlayer regions [21]. Additionally, K competes with Cs on the uptake carriers due to the similarity in chemical behavior; hence decreases Cs influx [9, 22]. Other elements, like Na, Ca, Al, P may decrease Cs availability to plant [23]. Moreover, increasing carbonate content in soil decreases the mobility of cesium [24]. Besides, organic matter can affect cesium availability [25]. In this concern, up to 71% of the ¹³⁷Cs was found bonded to organic matter in the forest of Japan after Fukushima accident [26]. Mostly, sorption of Cs on organic matter followed the Freundlich isotherm model [27].

Remediation of Cs-contaminated soils is therefore an obligation to ensure environmental safety [28-29]. This can be conducted via different techniques, such as bioremediation, thermal desorption electrokinetic remediation and soil leaching [30]. Chemical washing represents a good candidate for remediation of contaminants from soil via either acids (HNO₃, HCl, H₃PO₄ and H₂SO₄) [31] or synthetic chelators [32]. For example, the cationic surfactant (dodecyltrimethylammonium bromide (DTAB)) released sorbed cesium (up to 97%), in which ammonium ions substitute Cs within the interlayers of clay minerals [33]. Maybe, the usage of biodegradable components like NTA, EDDS, EDDG, EDDM, HIDS and PDA could be safer; yet still all of these agents exhibit relatively lower efficiencies in extraction of contaminants from soil versus EDTA-Na and DTPA [34] whose low molecular weights increase their efficiency to form strong complexes with Cs [35-36]. The current study aims at evaluating the efficiencies of both EDTA-Na and DTPA in washing Cs out of soils of different characters and varying contents of Cs (50, 100 and 200 mg Cs kg⁻¹) in relation to the risk assessment of cesium. Changes in different Cs fractions (pre- and post soil washing) is a matter of concern herein.

2. Materials and methods

2.1. Materials of study

Three surface soil samples (0-30 cm) were collected from different areas of Egypt, i.e. (S₁) and (S₂) from Mashtul El Souq and Anshas, respectively (Al-Sharqiyah Governorate) beside of (S₃) from Ameria industrial area, Alexandria Governorate. The first soil differs from the other ones in the textural class, the organic matter content

and the CaCO_3 content. The coordinates of the aforementioned sites are S1: 30° 21' 27" N and 31° 23' 02" E, S2: 30° 23' 19" N and 31° 26' 52" E and S3: 33° 17' 54" N and 44° 17' 59" E, respectively. These samples were air dried, crushed, finely ground, sieved through a 2 mm sieve, and analyzed for their chemical and physical characteristics according to Estefan et al. [37] and the results are presented in Table 1.

Soil samples were then artificially contaminated with non-radioactive (stable) Cs in the form of CsCl (sigma Aldrich, purity 99.9%) at three different rates i.e. 50, 100 and 200 mg kg^{-1} to simulate an accidental release of this contaminant to the surrounding soils. Soils were then left to equilibrate via subjecting to successive drying and wetting cycles for three months; thereafter soil portions were collected and analyzed for their content of Cs within the different fractions (sequential extractions) then subjected to washing with different chelating agents as revealed below.

2.2. Sequential extraction procedure

Subsamples of the contaminated soils underwent sequential fractionation to differentiate among different Cs fractions: water soluble, exchangeable, carbonate bound, Fe-Mn oxides bound, organic bound and residual fractions according to the procedures suggested by [38]. In this process, one gram portion of each of the previously artificially contaminated soils (on dry weight basis) was placed into a 40-mL polycarbonate centrifuge tube. The **water-soluble** fraction was extracted by **15 mL** of deionized water for 2 hours. **Eight mL** of 1 M MgCl_2 (pH = 7.0) were added to the residue from water-soluble fraction and agitated for one hour to extract the **exchangeable** fraction. The residue from exchangeable fraction was extracted by **8 mL** of sodium acetate (NaOAc) (adjusted to pH = 5.0) with continuous agitation for 5 hours to extract the **carbonate bound fraction**.

Concerning the Fe-Mn oxides-bound fraction, the residue from the carbonate fraction was mixed with **20 mL** of 0.04 M hydroxylamine hydrochloride ($\text{NH}_2\text{OH}\cdot\text{HCl}$) in 25% (v/v) acetic acid (HOAc) and heated up to 96°C with occasional agitation for 6 hours. For extracting the **organic-bound Cs fraction**, the residue from the previous step was extracted with 3 mL of 0.02 M HNO_3 and 5 mL of 30% H_2O_2 (adjusted to pH = 2.0 with HNO_3). This mixture was heated to 85°C for 2 hours, with occasional agitation. Furthermore, 3 mL aliquot of 30% H_2O_2 (adjusted to pH = 2.0 with HNO_3), was added and the mixture was heated again to 85°C for 3 hours with intermittent agitation. After cooling, 5 mL of 3.2 M NH_4OAc in 20% (v/v) HNO_3 was added and diluted to **20 mL** and agitated continuously for 30 minutes. The residual soil from the previous step was

placed in platinum crucible and digested with hydrofluoric acid to determine the residual Cs fraction.

Following each extraction procedure, mixtures were centrifuged at 5000 rpm for 10 minutes and the washing solutions were collected. All soil extractants were analyzed for Cs content using AAS technique then determined with Atomic Absorption Spectrometry AAS (model GBC 902). Total content of Cs was determined in soil via acid digestion via HF.

2.3. Washing the PTEs by chelating agents

Other portions of the contaminated soils were treated with either 0.05 M EDTA-Na or DTPA solution (pH adjusted to 7) at a rate of 1:10 (w/v: $W_{\text{soil}}/V_{\text{solution}}$) ratio, then left to equilibrate for 24 h via occasional shaking at room temperature. These mixtures were centrifuged, passed through a 0.45 μm filter and supernatants were collected. Thereafter, the soil samples were dried, and the sequential extraction of Cd was carried out again (after soil leaching via the synthetic chelating agents) as proposed by [38] to explore the redistribution of Cs among the different soil fractions.

Concentrations of Cs were determined in all liquid solutions obtained from the sequential extraction procedures and in the soil suspensions after EDTA-Na and DTPA washing. All extractions and digestions were performed in triplicate. Measurement of the metal ion contents in liquid extracts was determined with Atomic Absorption Spectrometry AAS (model GBC 902). All reagent used were of analytical grade.

2.4. Data analyses and calculations

The obtained data were statistically analyzed, using statistical software program SPSS Ver. 14 (IBM Co., USA) for ANOVA tests. Risk assessment code (RAC) was calculated as mentioned by [39] as follows:

$$\text{RAC} = \frac{\text{Ex} + \text{Carb}}{\text{Ex} + \text{Carb} + \text{Ox} + \text{OM} + \text{resid}} \times 100 \dots (1)$$

Where **Ex**: exchangeable fraction, **Carb**: carbonate bonded fraction, **Ox**: Fe-Mn oxide bonded fraction, **OM**: organic bonded fraction and **resid**: residual fraction. This equation has five classes based on RAC values which are no risk ($\text{RAC} < 1$), low risk ($1 \leq \text{RAC} < 10$), medium risk ($10 \leq \text{RAC} < 30$), high risk ($30 \leq \text{RAC} < 50$) and very high risk ($\text{RAC} \geq 50$). Many researchers use RAC for expressing the metal ion risk in soil and consider the water soluble fraction among the exchangeable fraction [40].

The removal efficiency (RE) of Cs by using the chelating agents was calculated from:

$$\text{Removal efficiency (RE)} = \frac{C_0 - C_w}{C_0} \times 100 \dots (2)$$

Where, C_0 is the C_s spiked level in the studied soil and C_w is the concentration of Cs remained in soil after washing using chelating agent.

Table (1) Some physical and chemical properties of the investigated soils.

Property	Soil location		
	Mashtoul El Souq	Anshass	Al-Amria
Clay %	28.8	9.30	12.60
Silt %	30.00	21.00	22.30
Fine Sand %	22.20	38.80	33.60
Coarse Sand %	19.00	30.90	31.50
Soil Texture	Clay Loam	Sandy loam	Sandy loam
Bulk density Mgm^{-3}	1.40	1.60	1.41
pH (1:2.5)	7.88	7.79	7.83
OM (g kg^{-1})	18	8.6	8.6
CaCO ₃ (g kg^{-1})	99	165	231
CEC (cmol kg^{-1})	24.32	4.05	10.97
EC (1:5) dS m^{-1}	2.49	5.89	3.29
	Soluble cations (mmol c L⁻¹)		
K ⁺	0.80	1.00	0.80
Na ⁺	13.50	31.00	16.50
Mg ⁺⁺	2.50	11.1	5.70
Ca ⁺⁺	7.20	14.9	9.00
	Soluble anions (mmol c L⁻¹)		
CO ₃ ⁻	ND	ND	ND
HCO ₃ ⁻	0.35	1.50	0.89
Cl ⁻	21.20	53.60	29.10
SO ₄ ⁻	2.45	2.90	2.01
	Cesium concentration (mg kg^{-1})		
Total Cs	ND	0.002	ND
Soluble Cs	ND	ND	ND

ND: Not Detected

3. Results and discussion

3.1. Effect of application of EDTA-Na or DTPA on washing of cesium from different tested spiked soils

Concentrations of Cs in the equilibrium solution after soil washing with either of the investigated chelating agents i.e. EDTA-Na or DTPA are presented in Figure 1. Both agents exhibited high efficiency to remove considerable concentrations of Cs from soils. In general, DTPA seemed to be more efficient in removal of Cs than EDTA-Na in all soils. These results contradict the outcomes of [32] who found that EDTA- Na was more effective than DTPA in washing soil cesium. Results also revealed that the removed amounts of Cs via soil washing were the highest

in Amreia soil (S_3), which contained the highest carbonate content, while the lowest ones were in Mashtoul El Souq soil (S_1) which had the highest organic matter content. These results probably indicate the capability of these chelating agents to chelate the carbonate bounded fraction of Cs while being incapable of chelating the organic bound fractions. In this concern, Cs is weakly bound to the carbonate fraction; thus can be extracted easily via chelating agents [41]; **yet**, organic matter has high affinity to sorb Cs [42]. **Generally**, the efficiency of chelates for washing Cs from soils is applicable at pH 4–8, as the EDTA- Na complexes can be re-adsorbed on soil surface sites at lower pH[43].

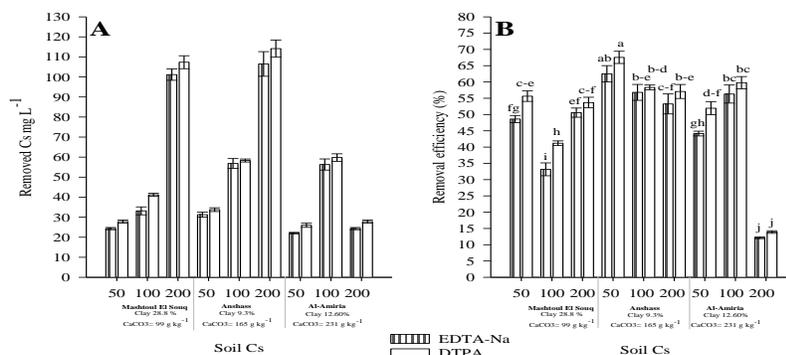


Fig. (1) Removal of Cs (mg/L)(means±standard deviations)from different soils spikedwith 50, 100 and 200mg Cs kg⁻¹via soil washing with EDTA-Na and DTPA (S1: Mashtoul El Souq soil, S2: Anshas soil and S3: Al- Ameria soil).

The removal efficiency of the tested chelators for Cs spiked soils were then calculated and the results are illustrated in Figure 2. The highest values were recorded for DTPA extraction versus EDTA-Na, particularly in S₂ (lowest clay and soil organic matter content), while being the least in S₁ which has the highest clay and organic matter contents. These results confirmed the high affinity of clay particles to retain Cs [44]as well as soil organic matter[45].

3.2. Effect of EDTA-Na and DTPA applications on Cs-fractions

Implications of soil washing with either EDTA or DTPA on different Cs fractions are represented in Figure

2. Generally, these chelates decrease considerably Cs contents within the different fractions, especially the water soluble, exchangeable and carbonate bond ones. In this concern, the efficiency of DTPA exceeded that of EDTA on decreasing water-soluble and exchangeable fractions of Cs regardless of the soil type or the initial Cs concentrations. It is thought that these two fractions are considered the mobile forms of Cs in soil.

Concerning the carbonate bound fraction of Cs, it was high in S₃ (soil of the highest CaCO₃) while exhibited comparable low (Anshas) versus the other two soil types.

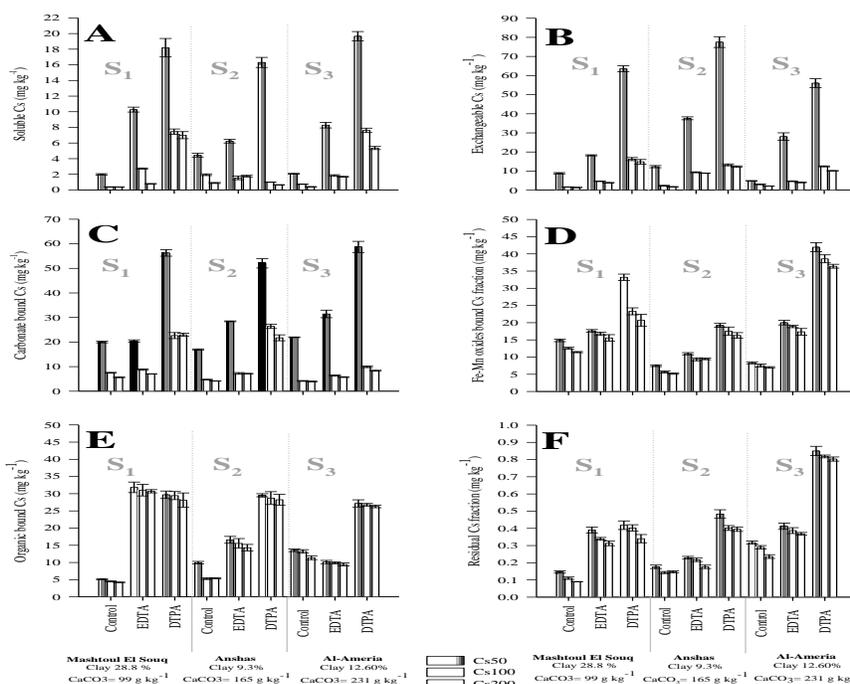


Fig. (2) Fractionation of cesium (means±standard deviations) of the different soils contaminated with 50, 100 and 200 mg Cs kg⁻¹(A: Water soluble, B: Exchangeable, C: Carbonate bond, D: Fe-Mn oxides bond, E: Organic matter bond and F: Residual fraction)

Variations within both the organic bound Cs and the residual fractions among the washed and unwashed soils were slight. Nevertheless, the residual fraction in all soils did not exceed 1 mg Cs kg⁻¹. May be this fraction was sorbed specifically on clay minerals, especially on the 2:1 clay mineral types, and this type of sorption is irreversible [46]. These clay minerals are characterized by their large surface area and high CEC,etc[19].

The total content of Cs in all soils is illustrated in Figure 3. Generally, DTPA decreased this content, especially in the sandy loam soil of relatively low CaCO₃ content whereas the lowest effect appeared due to EDTA-Na application on the clayey soil. Overall, the chelating agents can mobilize effectively metal contaminants [47].

3.3. Effect of EDTA-Na and DTPA application on risk assessment code

The risk assessment code of Cs was calculated in spiked soils (pre and post soil washing with either EDTA-Na or DTPA) and the obtained results are shown in Fig 3 and Table 2. Overall, the pre-washing value signify very high risk (RAC >50) except for the heavily textured soil (the clayey) which was contaminated with 100 mg Cs kg⁻¹ as it exhibited only high Cs risk (49.54). DTPA was, generally, more effective in decreasing RAC than EDTA-Na. After soil washing, the risk assessment code decreased noticeably from very high to medium. Such reductions seemed to be less detectable in the sandy loam soil which exhibited lower CaCO₃ content (Anshass) versus the other two soils. These results therefore agree with that mentioned by [30].

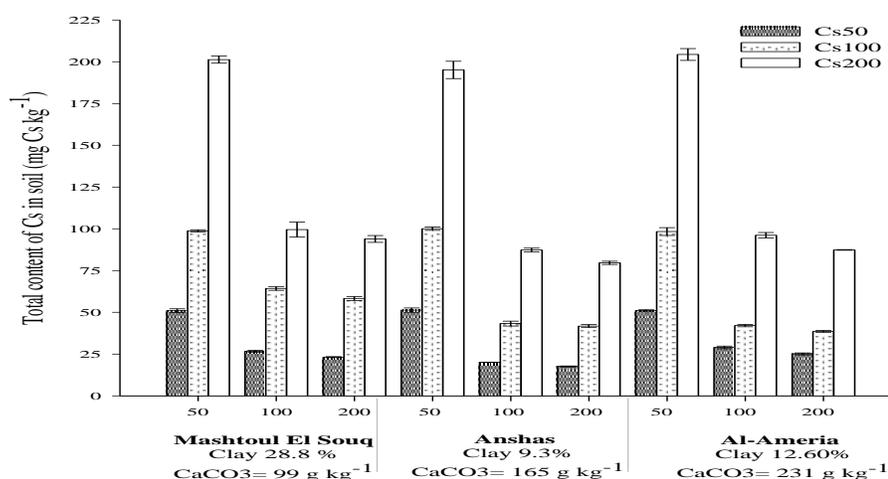


Fig. (3) Effect of washing agents (EDTA-Na and DTPA) on total Cs content (means±standard deviations) in spiked soils

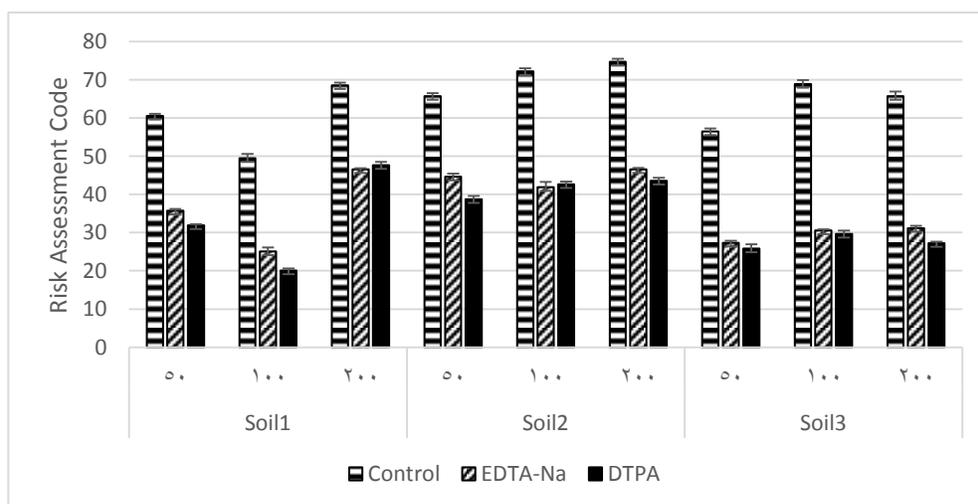


Fig. (4) Risk assessment code (RAC) (means±standard deviations) for soil washed with chelators (EDTA-Na and DTPA)

Table (2) Risk assessment code (RAC) (means±standard deviations)for soil washed with chelators (EDTA-Na and DTPA) .

		Initial Cs (mg kg ⁻¹)			
		Cs50	Cs100	Cs200	Mean
Soil1	Pre-soil washing	60.541±0.566	49.543±1.012	68.559±0.648	59.547
	Post- EDTA wash	35.792±0.356	25.153±0.941	46.595±0.278	35.847
	Post DTPA wash	31.973±0.23	20.079±0.517	47.69±0.81	33.247
	Mean	42.769	31.591	54.281	42.880
Soil2	Pre-soil washing	65.702±0.756	72.249±0.729	74.746±0.767	70.899
	Post DTPA wash	44.711±0.694	41.945±1.325	46.577±0.315	44.411
	Post DTPA wash	38.801±0.759	42.669±0.678	43.594±0.764	41.688
	Mean	49.738	52.288	54.973	52.333
Soil3	Pre-soil washing	56.519±0.714	68.924±0.942	65.763±1.155	63.735
	Post DTPA wash	27.342±0.533	30.589±0.304	31.218±0.585	29.717
	Post DTPA wash	25.85±1.121	29.7±0.84	27.286±0.289	27.612
	Mean	36.571	43.071	41.422	40.355

Conclusion

Soil washing with DTPA was more efficient in extracting Cs from contaminated soils versus EDTA-Na. Water-soluble, exchangeable and carbonate Cs fractions decreased considerably owing to the leaching protocol, while the organic-bond and residual fractions slightly changed. Mostly soil properties affected extensively the removal efficiency of Cs within the different fractions. In particular, clay and CaCO₃ contents decreased significantly this efficiency. Risk assessment code, which depends on the change of water soluble, exchangeable and carbonate fractions, decreased from very high risk to medium risk for most of the studied soils after washing contaminated soils with the investigated chelating agents. Accordingly, these results guarantee the usage of DTPA and EDTA-Na as efficient chelating agents for soil washing of cesium from contaminated soils.

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