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The influence of particle size on radionuclide activity concentrations in Tejo River sediments

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ABSTRACT

Sediment samples from Tejo River were analyzed for ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K by HPGe gamma spectrometry. The activity concentration data were statistically analyzed. The activity concentrations values were in the range of about two orders of magnitude for each radionuclide. The influence of the particle size on the radionuclide concentrations was observed. The different environmental origins of the radionuclides ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K, in the sediments were demonstrated through correlation analysis. Cluster analysis showed a close relationship between ^{228}Ra and ^{226}Ra and a different behavior for ^{40}K. The data obtained in this study provides useful information on the background radioactivity of the studied area and can be further used for radiological mapping of the Tejo River.

1. Introduction

Radioactivity occurs naturally in the environment. The radionuclide content of a river depends on mineralogical features in the catchment area and the chemistry of the river as a whole. The major sources of natural radionuclides in sediments have different possible origins. These include weathering and recycling of terrestrial minerals and rocks (igneous or metamorphic) containing ^{40}K and radionuclides of the uranium and thorium radioactive series, rainfall and other depositional phenomena such as gravitational settling and precipitation. In stream sediments U and Th may be found incorporated into the existing minerals or they may be adsorbed directly from river water onto clay minerals or organic debris. Occasionally, U may be removed from river water to sediments directly if reducing bottom conditions exist (El-Gamal et al., 2007).

Artificial radionuclides can be introduced into rivers for direct and indirect inputs. Directly through the aqueous discharges from nuclear installations and indirectly from wash-off of land deposited activity within the river catchment following nuclear weapon testing or nuclear accidents. Most of the radioactivity deposited on surface soils, depending on the radionuclide's geochemistry, is washed out by rains and drained in to the rivers. After reaching the river ecosystem, radionuclides may be transferred through the water-sediment-biota pathways (Ajayi and Kuforiji, 2001; Ajayi, 2008) to humans, by using the river water as drinking water or for irrigation and by the consumption of contaminated fish.

River sediments, consisting of mineral particles with different sizes, are considered long term reliable indicators of river pollution by radionuclides because water pollution components are deposited in the sediments. Long-term radioactive pollution may accumulate and whenever the sediments are re-suspended the radionuclides re-enter the sediment-biota chain (Bikit et al., 2006). Therefore, the knowledge of the concentrations and distribution of the radionuclides in the river sediments are of great interest since it provides useful information on the background and on the temporal changes in radionuclide activity concentrations within the river. In the framework of the environmental radioactivity survey performed in Portugal (Madruga, 2008) several components of the Tejo River, including sediments, have been monitored (Madruga et al., 2009). The Tejo River originating in Spain runs through Portugal on its way into the Atlantic Ocean. The Tejo River receives discharges from three Spanish Nuclear Power Plants (NPP) with the Almaraz NPP located nearest the Portuguese border (about 120 km). In Portugal, the sedimentation load and natural distribution of sediments are affected by two dams (Fratel and Belver) (Carreiro and Sequeira, 1998).

The main objective of this study is to assess, through statistical analysis, the activity distribution of natural (^{228}Ra, ^{226}Ra, ^{40}K) and anthropogenic (^{137}Cs) radionuclides in sediments collected in Tejo

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River and the influence of the sediment particle sizes on radionuclides concentrations. Besides, the knowledge of radionuclides concentrations and distribution in Tejo River sediments can be useful for the radiological mapping of the studied area.

2. Material and methods

2.1. Sampling and sample preparation

Surface sediment samples, around 4 kg each, were collected monthly (from years 2001–2009) in Tejo River at two selected locations: Vila Velha de Ródão (VVR) near the Spanish border and upstream of Fratel and Belver dams, and Valada (V) downstream of these dams (Fig. 1).

The samples were collected with a metal sampler (Berthois cone) and stored into labeled plastic boxes. At the laboratory the samples were homogenized, dried at room temperature, crushed, sieved and fractions lower than 1000 μm (*A*-bulk sediment), between 250 μm and 63 μm (*B*-sand) and lower than 63 μm (*C*-silt/clay) were used for analysis. Representative aliquots of each sieved sample were transferred to 180 cm^3 or 10 cm^3 plastic containers (depending on the amount available), closed, sealed and left aside for about one month to ensure that secular equilibrium is reached between radium isotopes and its short lived progeny in uranium and thorium radionuclide series.

2.2. Gamma spectrometry analysis

Samples were counted for 15 h and analyzed for ^{228}Ra (determined through ^{228}Ac), ^{226}Ra (determined through ^{214}Pb and ^{214}Bi daughters), ^{137}Cs and ^{40}K by gamma spectrometry.

The measurement system is composed by HPGe detectors and standard analog electronics. The detectors were shielded from environmental background, by using lead shields with copper and tin lining. The efficiency calibration was performed using NIST-traceable multinuclide radioactive standards, covering the energy range 46.5–1836 keV, customized to reproduce the exact geometries of the samples in a water-equivalent epoxy resin matrix by Analytics, Incorporated. The density correction was carried out through GESPECOR version 4.2 software® (Sima et al., 2001) and Genie 2000® (Canberra) was used for data acquisition and analysis. The stability of the system (activity, FWHM, centroid) was

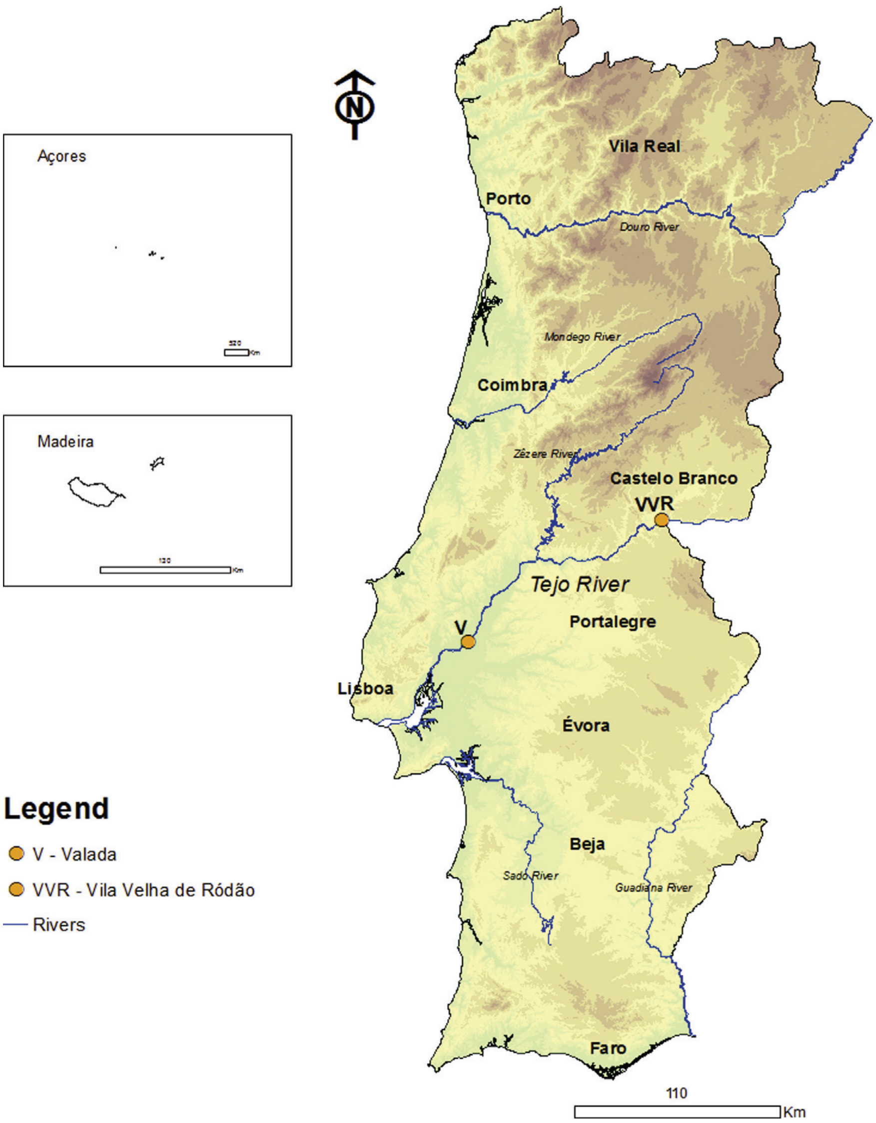

Fig. 1. Sampling locations in Tejo River (Portugal).

Table 1
Statistical parameters of ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K (Bq kg^{-1}, dry weight) distribution in sediment samples from Tejo River.

Radionuclide	Size particle	Vila Velha de Ródão							Valada						
		Mean (*N*)	Median	Sdev	Mad	Range	S–W Orig	S–W log	Mean (N)	Median	Sdev	Mad	Range	S–W Orig	S–W log
^{228}Ra	*A*	68 (101)	62	37	22	11–251	*0.000*	0.405	32 (105)	29	15	8	12–83	*0.000*	0.081
	B	185 (101)	145	192	67	32–1583	*0.000*	0.159	68 (105)	63	28	16	18–155	*0.000*	0.253
	C	295 (101)	254	160	77	41–802	*0.000*	0.099	236 (105)	198	149	54	74–1131	*0.000*	*0.015*
^{226}Ra	*A*	43 (101)	41	17	10	20–124	*0.000*	0.115	22 (105)	21	8.8	6	11–50	*0.000*	*0.033*
	B	95 (101)	77	81	27	27–724	*0.000*	*0.008*	42 (105)	40	15	9	14–84	*0.008*	0.489
	C	180 (101)	167	85	42	30–479	*0.000*	0.189	144 (105)	121	81	30	60–549	*0.000*	*0.001*
^{137}Cs	*A*	1.7 (94)	1.6	1.0	0.3	0.3–7.6	*0.000*	*0.000*	0.9 (78)	0.8	0.4	0.2	0.3–2.9	*0.000*	0.637
	B	2.2 (81)	1.9	1.1	0.4	0.7–8.7	*0.000*	*0.003*	1.2 (74)	1.1	0.6	0.3	0.3–3.7	*0.000*	0.100
	C	6.2 (79)	5.8	2.2	1.4	1.7–12.9	0.140	*0.027*	4.0 (62)	3.9	1.7	1.1	1.5–11.9	*0.000*	0.280
^{40}K	*A*	890 (101)	886	78	46	645–1106	*0.045*	0.083	882 (105)	881	82	44	492–1068	*0.000*	*0.000*
	B	769 (101)	774	88	42	333–1101	*0.000*	*0.000*	842 (105)	829	77	37	664–1283	*0.000*	*0.000*
	C	681 (101)	675	105	61	382–1096	*0.042*	*0.015*	807 (105)	807	114	57	467–1271	*0.003*	*0.000*

A – sediment size particle <1000 μm; *B*, sediment size particle between 250 μm and 63 μm; *C*, sediment size particle <63 μm; Mad- median absolute deviation.
S–W Orig and S–W log represent the *p* values for the Shapiro–Wilk test for normal distribution of the untransformed (original) and log-transformed data respectively; The *p* values <0.05 are indicated in italics.

monitored at least once a week with a ^{152}Eu point source. The quality of the measurements was also demonstrated through the participation in intercomparison exercises organized by international organizations (Romero et al., 2006; IAEA-CU-2006-04, 2007; IAEA-CU-2007-4 2009).

3. Results and discussion

The quantified results on radionuclide activity concentrations in sediment samples were statistically analyzed using the STATISTICA 12 package®.

Table 1 presents the mean, median, standard deviation (Sdev), median absolute deviation (Mad) and the range of the ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K activity concentrations values for the Tejo River sediment samples. Regarding the ^{137}Cs, only the values higher than the minimum detection activity were considered in this study.

A dispersion of activity concentrations was observed for all the radionuclides with values ranging from 1 to 2 orders of magnitude which were corroborated by the large difference between median absolute deviation values. The median absolute deviation gives an absolute measure of dispersion that was not affected by extreme outliers and by the standard deviation. For example, values of 67 Bq kg^{-1} and 77 Bq kg^{-1} for Mad and 192 Bq kg^{-1} and 160 Bq kg^{-1} for Sdev respectively were obtained for ^{228}Ra in grain size fractions B and C at Vila Velha de Ródão. As it is shown in Table 1 the mean values were higher than the median values indicating a positive (to the right) skewed distribution. The frequency distributions of ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K activity concentrations were considered separately for each sediment fraction and analyzed for the original concentration activity data applying the Shapiro–Wilk test for normal distribution. The S–W test result, *p* value, is the probability which can support a decision as to whether the null hypothesis is to be rejected. If *p* value is smaller than the predefined significance level (0.05) the null hypothesis has to be rejected (Reimam et al., 2008). The *p* value obtained for the original data set (Table 1) were lower than 0.05 (*values in italics*), with exception for the ^{137}Cs distribution in silt/clay fraction of VVR sediments, showing that the data do not follow a normal distribution as expected, since the mean and median values were not close. It was reported that (Haynes, 1996) when the logarithms of positively skewed observations are plotted in a histogram the result is frequently near to a normal distribution and the data are said to have a log-normal distribution. Table 1 showed the *p* values for the Shapiro–Wilk test of the log-transformed data. The distribution patterns present, for some variables, a *p* value higher than the chosen significance level (0.05) and, in these conditions, it can be accepted that these data followed a lognormal distribution. As an example, an illustration of the ^{228}Ra and ^{137}Cs log data distribution in VVR and V were presented in Figs. 2 and 3 respectively. As reported in the literature (Reimam and Filzmoser, 1999) in the majority of cases the data transformation (e.g. log, ln) does not result necessarily in a normal distribution.

In order to know the relationships between the measured variables, Spearman correlation coefficients (r_s), a nonparametric parameter which measure the strength of association between two ranked variables, were computed. The values presenting a significance level of 95% ($p < 0.05$) for each radionuclide and for each sediment fraction were shown in Tables 2 and 3 for Vila Velha de Ródão and Valada respectively.

Regarding Vila Velha de Ródão (Table 2), positive correlations were observed between ^{228}Ra and ^{226}Ra for all sediment fractions. To give one idea of the variables dependence, the Spearman rank correlations between the ^{228}Ra and ^{226}Ra activity concentrations for sediment fraction C and between the ^{226}Ra activity concentrations for sediment fraction A and B were shown in Figs. 4 and 5 respectively. The higher correlation coefficients (ranging from 0.90 to 0.97) obtained between both radionuclides for each fraction

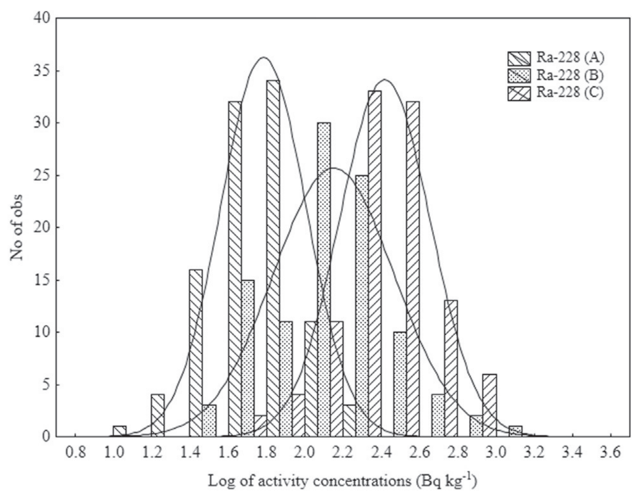

Fig. 2. Distribution of ^{228}Ra (sediment grain fractions A, B and C) in Vila Velha de Ródão.

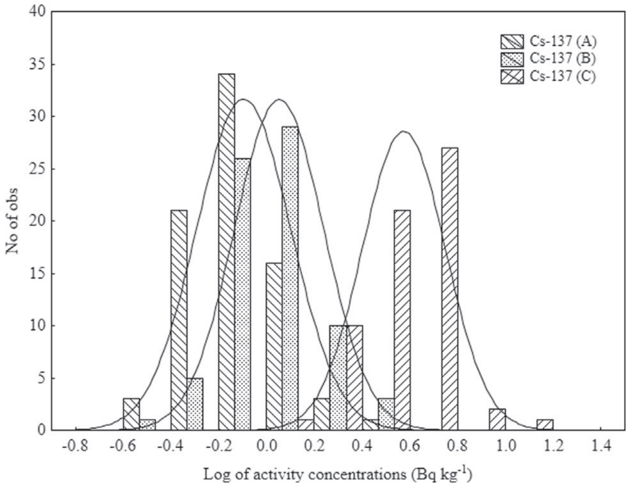

Fig. 3. Distribution of ^{137}Cs (sediment grain fractions A, B and C) in Valada.

(A, C and B) indicated that ^{228}Ra and ^{226}Ra had the same geological origin, as these radionuclides are associated to specific mineralogical components. Moreover, the strong correlations between these radionuclides imply that they are similarly affected by transport and by physical and chemical interaction processes in the aquatic environment. The ^{137}Cs and ^{40}K activity concentrations were in general not correlated with the other radionuclides (Table 2). This finding could be related with the radionuclides distribution in the sediments bed. The natural radionuclides are integral part of the minerals structure that constitutes the sediments while the ^{137}Cs was distributed in the environment from fallout due to the atmospheric testing of nuclear weapons in the 1960s and to the Chernobyl accident (Van der Graaf et al., 2007). In general, a behavior similar to those obtained for Vila Velha de Ródão was observed for the ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K activity concentrations relationships regarding the data from Valada (Table 3, Figs. 6 and 7).

For both locations (VVR and V) the stronger correlation coefficients between ^{228}Ra and ^{226}Ra for the same sediment grain size should be emphasized when compared with the relationship between all sediment fractions for the same radionuclide (Tables 2and 3). Considering the same radionuclide, better correlations for fractions A and B were obtained than for fractions A and C. This is in agreement to what may be expected since the fraction B (sand), representing 28.7% of the fraction A (*bulk* sediment) in VVR and 24.1% in V, might have a geological and mineralogical composition more similar to the bulk sediment than the fraction C (silt/clay) consisting in a very small percentage of the fraction A, 1.8% and 2.2% in VVR and V respectively.

In both sampling sites (VVR and V) the mean and median activity concentrations for ^{228}Ra were higher than for ^{226}Ra (Table 1) due to the low geochemical mobility and insoluble nature in water of ^{232}Th (^{228}Ra progeny) when compared to ^{238}U (^{226}Ra progeny).

It is well known that the adsorption of radioactive elements has a strong dependence on the nature of sediment and decreases from

Table 2
Spearman rank order correlations (r_s values, significance level $p < 0.05$) for sediments from Vila Velha de Ródão.

		^{228}Ra			^{226}Ra			^{137}Cs			^{40}K		
		A	*B*	*C*	*A*	*B*	*C*	*A*	*B*	*C*	*A*	*B*	*C*
^{228}Ra	*A*		0.72	0.28	0.90	0.72	0.20	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
	B	0.72		0.27	0.60	0.97	0.20	n.s.	−0.24	0.25	n.s.	n.s.	n.s.
	C	0.27	0.27		n.s.	0.22	0.95	−0.23	−0.31	n.s.	n.s.	n.s.	n.s.
^{226}Ra	*A*	0.90	0.60	n.s.		0.66	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
	B	0.72	0.97	0.22	0.66		n.s.	n.s.	−0.22	0.25	n.s.	n.s.	0.20
	C	0.20	0.20	0.95	n.s.	n.s.		−0.22	−0.28	n.s.	n.s.	n.s.	n.s.
^{137}Cs	*A*	n.s.	n.s.	−0.23	n.s.	n.s.	−0.22		0.60	n.s.	0.26	n.s.	−0.33
	B	n.s.	−0.24	−0.31	n.s.	−0.22	−0.28	0.60		0.33	n.s.	n.s.	−0.32
	C	n.s.	0.25	n.s.	n.s.	0.25	n.s.	n.s.	0.33		n.s.	n.s.	n.s.
^{40}K	*A*	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	0.26	n.s.	n.s.		0.42	n.s.
	B	−0.21	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	0.42		n.s.
	C	n.s	n.s.	n.s.	n.s.	0.20	n.s.	−0.33	−0.32	n.s.	n.s.	n.s.	

A – sediment size particle <1000 μm; *B*- sediment size particle between 250 μm and 63 μm; *C*- sediment size particle <63 μm.
n.s. – not significant ($p > 0.05$).

Table 3
Spearman rank order correlations (r_s values, significance level $p < 0.05$) for sediments from Valada.

		^{228}Ra			^{226}Ra			^{137}Cs			^{40}K		
		A	*B*	*C*	*A*	*B*	*C*	*A*	*B*	*C*	*A*	*B*	*C*
^{228}Ra	*A*		0.53	0.53	0.96	0.57	0.53	0.56	n.s.	n.s.	n.s.	−0.43	n.s.
	B	0.53		0.39	0.45	0.94	0.37	n.s.	n.s.	n.s.	n.s.	−0.22	n.s.
	C	0.53	0.39		0.43	0.32	0.94	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.
^{226}Ra	*A*	0.96	0.45	0.43		0.51	0.44	0.62	n.s.	−0.27	n.s.	−0.41	n.s.
	B	0.57	0.94	0.32	0.51		0.33	n.s.	n.s.	n.s.	n.s.	−0.24	n.s.
	C	0.53	0.37	0.94	0.44	0.33		n.s.	n.s.	n.s.	n.s.	−0.20	n.s.
^{137}Cs	*A*	0.56	n.s.	n.s.	0.62	n.s.	n.s.		0.38	n.s.	n.s.	−0.29	−0.28
	B	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	0.38		0.34	n.s.	n.s.	n.s.
	C	n.s.	n.s.	n.s.	−0.27	n.s.	n.s.	n.s.	0.34		n.s.	n.s.	0.30
^{40}K	*A*	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.		0.34	n.s.
	B	−0.43	−0.22	n.s.	−0.41	−0.24	−0.20	−0.29	n.s.	n.s.	0.34		n.s.
	C	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	−0.28	n.s.	0.30	n.s.	n.s.	

A – sediment size particle <1000 μm; *B*- sediment size particle between 250 μm and 63 μm; *C*- sediment size particle <63 μm.
n.s. – not significant ($p > 0.05$).

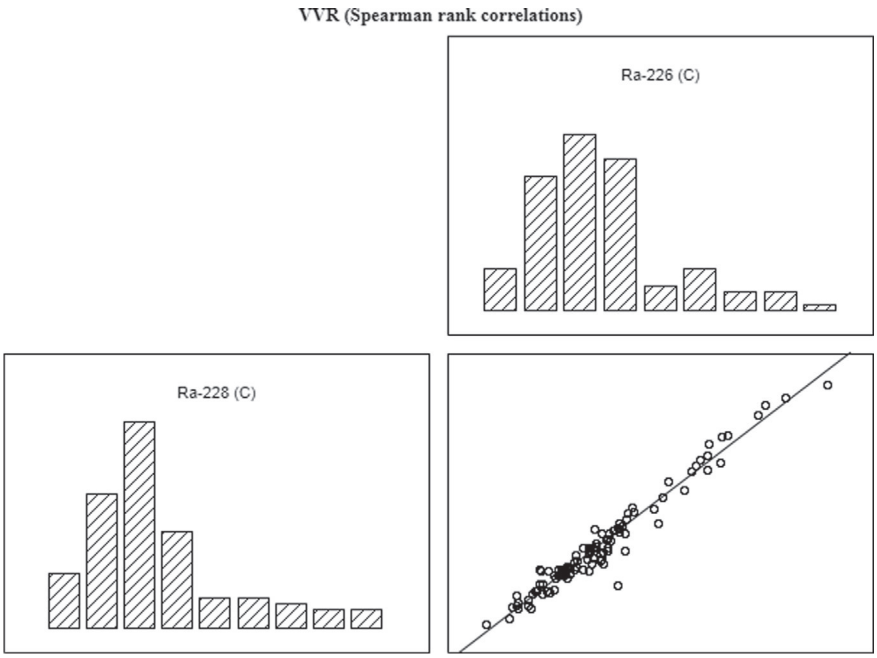

Fig. 4. Spearman rank correlations of ^{228}Ra and ^{226}Ra (sediment grain fraction C) in Vila Velha de Ródão.

fine to coarse-grained materials. According to McCubbin et al. (2004) the highest concentrations of most radionuclides in bulk Irish Sea sediments are associated with fine grain materials. In general, the U–Th radionuclides are associated with heavy minerals whereas ^{40}K is concentrated within clay minerals (Tsabaris et al., 2007; Rodríguez et al., 2008). Indeed, regarding the influence of the sediment particle size, Table 1 showed an increase in the mean and in the median activity concentration values with decreasing particle size for ^{228}Ra, ^{226}Ra and ^{137}Cs, seemed to indicate that fractions A, B and C are mineralogical different and consequently have different radionuclides contents. However, a slight decrease in ^{40}K is observed contrarily to what is expected, probably due to the existence of lower amount of feldspars, richer in potassium, in the silt/clay fraction (C) when compared with the other fractions.

The mean activity radionuclide concentration values in Tejo River sediments (Table 1) were in the same order of magnitude of natural radionuclide content in Portuguese soils (UNSCEAR, 2000) and higher than the worldwide average concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K reported by UNSCEAR (2000) which are 35, 30 and 400 Bq kg^{-1}, respectively.

The mean concentration activities data for all the radionuclides were analyzed using the *t*-test for independent samples. Considering the dispersion of the data distribution, the test was applied to

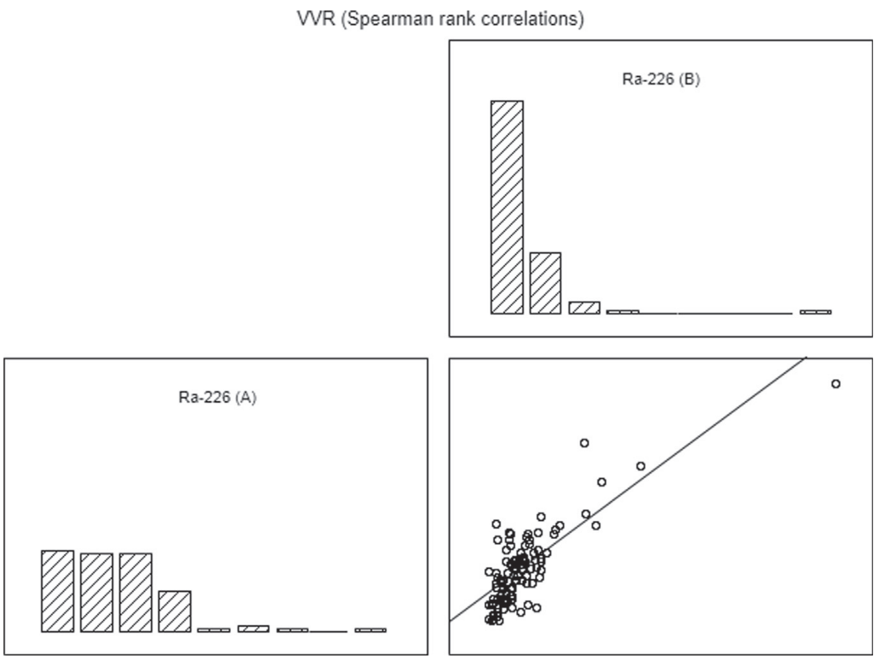

Fig. 5. Spearman rank correlations of ^{226}Ra (sediment grain fractions A and B) in Vila Velha de Ródão.

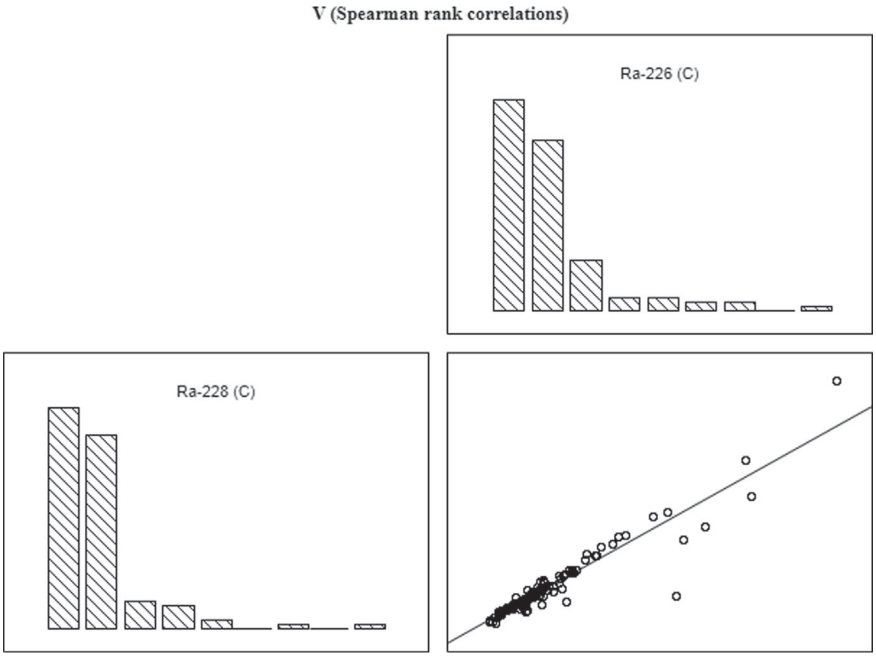

Fig. 6. Spearman rank correlations of ^{228}Ra and ^{226}Ra (sediment grain fraction C) in Valada.

the log data. Fig. 8 showed the log of the activity concentration for the different size fractions for all the radionuclides and for the two locations. The activity concentration values for both locations (VVR and V) and for each radionuclide were significantly different, at 95% confidence level ($p < 0.05$), for the different sediment fractions with exception of: ^{228}Ra (fraction B) VVR vs ^{226}Ra (fraction C) VVR; ^{228}Ra (fraction A) VVR vs ^{228}Ra (fraction B) V; ^{228}Ra (fraction B) VVR vs ^{226}Ra (fraction C) V; ^{226}Ra (fraction A) VVR vs ^{226}Ra (fraction B) V; ^{40}K (fraction A) VVR vs ^{40}K (fraction A) V. This is in agreement with the different geological composition of the two regions which are mainly constituted by outcrops of metamorphic, sedimentary and igneous rocks in VVR and by detrital sedimentary formations in V (Ferreira, 2000).

The data were also analyzed through a hierarchical cluster analysis by using STATISTICA 12 package®. This multivariate statistical technique involves splitting a data set into a number of groups of observations allowing a statistical identification of relatively homogeneous sample groups based on the radionuclide activities. This is an exploratory data analysis tool that can be used to unveil structures in data without providing an explanation or interpretation (Lambrakis et al., 2004; Külahci and Sen, 2008; Suresh et al., 2012). The orders in which parameters or variables

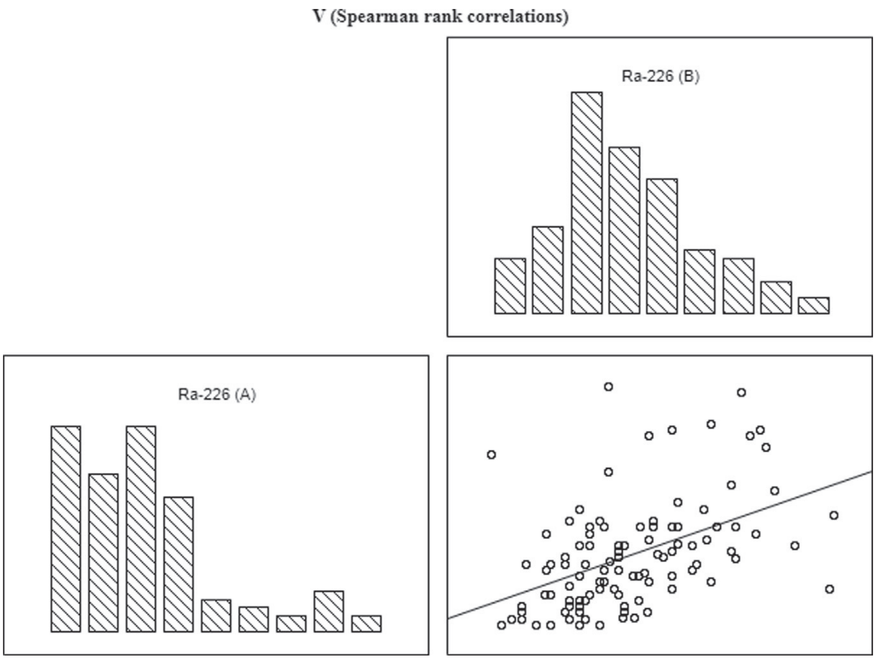

Fig. 7. Spearman rank correlations of ^{226}Ra (sediment grain fractions A and B) in Valada.

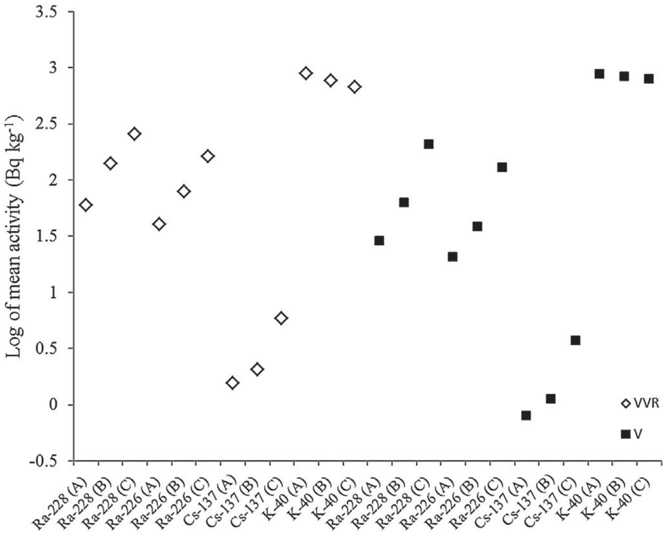

Fig. 8. The ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K log mean activities for A, B and C sediment grain fractions in Vila Velha de Ródão (VVR) and Valada (V).

are combined to form clusters with similar properties are visually displayed as dendrograms.

The dendrograms obtained for the two sampling sites and for the natural radionuclides, ^{40}K, ^{226}Ra and ^{228}Ra were presented in Fig. 9. The ^{137}Cs is not considered in this analysis due to its low concentration values when compared with the other radionuclides. Two main groups of samples were distinguished: one formed by the natural radionuclide ^{40}K and the other by the radionuclides of the U and Th series, ^{226}Ra and ^{228}Ra. The cluster analysis showed the close relationship between the two radionuclides of the U and Th series (supporting what was presented above) and separates ^{40}K in a group far from the others which is related to the different geological origin and geochemical behavior of the radionuclides. Regarding the ^{40}K, two sub-groups were formed with the lower particle size well distinguished suggesting a different mineralogical composition.

The activity concentration ratio ^{228}Ra/^{226}Ra was also calculated for all samples. The mean values obtained presented in Table 4 ranged from 1.53 to 1.83 at Vila Velha de Ródão and from 1.41 to 1.62 at Valada. The ^{228}Ra/^{226}Ra mean and the respective standard

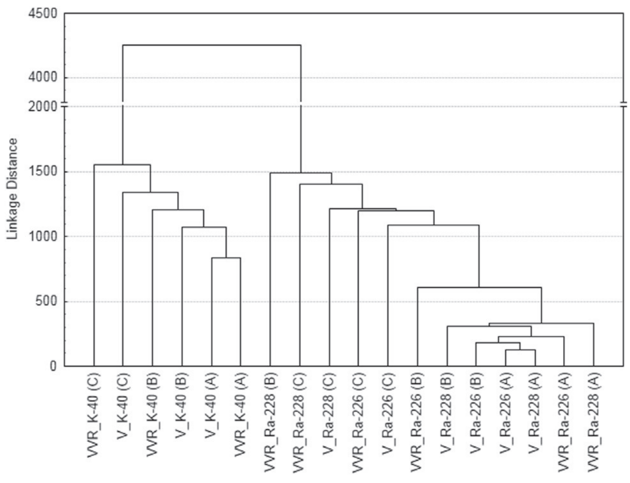

Fig. 9. Dendrogram showing groups among the radionuclides ^{228}Ra, ^{226}Ra and ^{40}K for A, B and C sediment grain fractions in Vila Velha de Ródão (VVR) and Valada (V).

Table 4
The ^{228}Ra/^{226}Ra ratio values for sediments samples from Tejo River.

Sed. size particle	Vila Velha Ródão			Valada		
	Mean (N)	Std. Dev.	Range	Mean (N)	Std. Dev.	Range
A	1.53 (101)	0.30	0.26–2.02	1.41 (105)	0.20	0.93–2.25
B	1.83 (101)	0.62	0.46–7.1	1.59 (105)	0.21	1.00–2.14
C	1.60 (101)	0.22	0.81–2.18	1.62 (105)	0.22	0.60–2.06

A – sediment size particle <1000 μm; *B*, sediment size particle between 250 μm and 63 μm; *C*, sediment size particle <63 μm.

deviation values seemed to show that the ratios do not depend of the grain size fractions. The ratio values higher than 1 could be related, as already mentioned, to the lower geochemical mobility and insoluble nature of thorium in water when compared to uranium.

4. Conclusions

This study of ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K distribution in Tejo River sediments demonstrates the influence of the grain size on the radionuclides activity concentrations in river sediments. An increase (^{228}Ra, ^{226}Ra, ^{137}Cs) and a decrease (^{40}K) of the activity concentrations with the decreasing sediment particles size were found. The different origins of the radionuclides in the sediments, resulting from their mineralogy (^{228}Ra, ^{226}Ra and ^{40}K) or from fallout (^{137}Cs) were indicated by the correlation analysis. The cluster analysis for ^{228}Ra, ^{226}Ra and ^{40}K, distinguished two groups, one constituted by the uranium and thorium radionuclides series (^{228}Ra, ^{226}Ra) and another one by the natural radionuclide, ^{40}K. This fact can be attributed to the different geological formations to which the radionuclides are associated. These findings and follow up research are expected to contribute to the radiological mapping of the Tejo River.

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