Journal of Sustainable Mining 18 (2019) 45–51

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Journal of Sustainable Mining

journal homepage: www.elsevier.com/locate/jsm

Research paper

Natural radionuclides (NORM) in a Moroccan river affected by former conventional metal mining activities

Guillermo Manjón[a], Juan Mantero[a], Ignacio Vioque[a,*], Inmaculada Díaz-Francés[a], José Antonio Galván[a], Said Chakiri[b], Abdelmajid Choukri[c], Rafael García-Tenorio[a,d]

[a] Applied Nuclear Physics Group, Applied Physics II Department, ETSA, Universidad de Sevilla, Av Reina Mercedes 2, 41012, Seville, Spain
[b] Geosciences of Natural Resources Laboratory, IbnTofail University, Faculty of Sciences, BP 133, 14000, Kénitra, Morocco
[c] Laboratory of Polymers, Radiations and Environment, IbnTofail University, Faculty of Sciences, BP 133, 14000, Kénitra, Morocco
[d] Spanish National Accelerator Center (CNA), Universidad de Sevilla-Junta de Andalucía-CSIC, Seville, Spain

ARTICLE INFO

Keywords:
NORM (Naturally occurring radioactive material)
Abandoned mines
Moulouya river
Morocco

ABSTRACT

The main aim of this work was to determine the levels of multiple natural radionuclides in an aquatic system (Moulouya river, Morocco) impacted by multiple abandoned Zinc and Lead mines. ^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{210}Po were determined by alpha-particle spectrometry in water and sediment samples collected along the river and in samples from three pit lakes of abandoned mines, located in the Upper Moulouya catchment area.

The results enabled the analysis of the different levels of impact of former mining activities, depending on the natural radionuclides. While the activity concentration of U-isotopes in Moulouya river water was slightly elevated in the vicinity of abandoned mine wastes, other natural radionuclides (Th-isotopes and ^{210}Po) levels were typical of a natural environment. This fact is clearly reflected in the magnitude and range observed in the distribution coefficients for the different radionuclides analyzed.

1. Introduction

Natural radioactivity is omnipresent in the Earth's environment. All living beings are exposed to ionizing radiation and it is well-known that human activities such as the mining and processing of some raw materials can increase both the aforementioned exposure and the concentration of natural radionuclides in the environment. While it is true that occupational risks due to these elevated concentrations have been deeply studied and even regulated, their environmental role has not been given much consideration. The importance of measuring the radioactive environmental consequences of non-nuclear industry and mining, not related to nuclear-fuel cycle, was recently highlighted by the International Commission of Radiation Protection (ICRP) (Michalik, Brown, & Krajewski, 2013).

Generate waste material from mining activities can be potentially hazardous to the environment. For this reason, mining corporations need to prioritize their management in order to prevent the contamination of ground water, rivers and lakes (Kossoff et al., 2014). The waste formed can be, in some specific cases, a source of natural radionuclides to the environment and the analysis of their behavior is imperative in order to minimize its impact.

Although there are many cases of studies on rivers affected by uranium mines (Carvalho, Oliveira, Lopes, & Batista, 2007), the NORM environmental effects of non-uranium mines on aquatic systems are rarely mentioned in the literature, with a few exceptions (Barbero et al., 2014; Huang et al., 2015). Natural radioactivity in different ecosystems, which are potentially affected by non-uranium mining, are often excluded from radiological control due to the moderate levels of radioactivity, but radionuclides can play a role as tracers of metal and other anthropogenic contaminations. Some recent articles describe aquatic systems where both natural radionuclides and heavy metal concentrations have been used as tracers (González-Labajo, Bolívar, & García-Tenorio, 2001; Saç, Ortabuk, Kumru, Içhedef, & Sert, 2012).

In this paper, the objective is to evaluate the possible NORM impact generated by the legacies remaining from former Lead and Zinc mining activities in the catchment area of the Moulouya river (Morocco) and to study the different behavior of the natural radionuclides analyzed once they have been incorporated in to the aquatic system.

2. Materials and methods

The Upper Moulouya Lead district, Fig. 1, is crossed by the Moulouya river (Oued Moulouya), which is the most important river in Morocco. The geological and geographical description of the area is

* Corresponding author.Departamento de Física Aplicada II, Universidad de Sevilla, E-41012, Sevilla, Spain.
E-mail address: ivioque@us.es (I. Vioque).

https://doi.org/10.1016/j.jsm.2019.02.003
Received 26 December 2018; Received in revised form 18 January 2019; Accepted 12 February 2019
Available online 15 February 2019

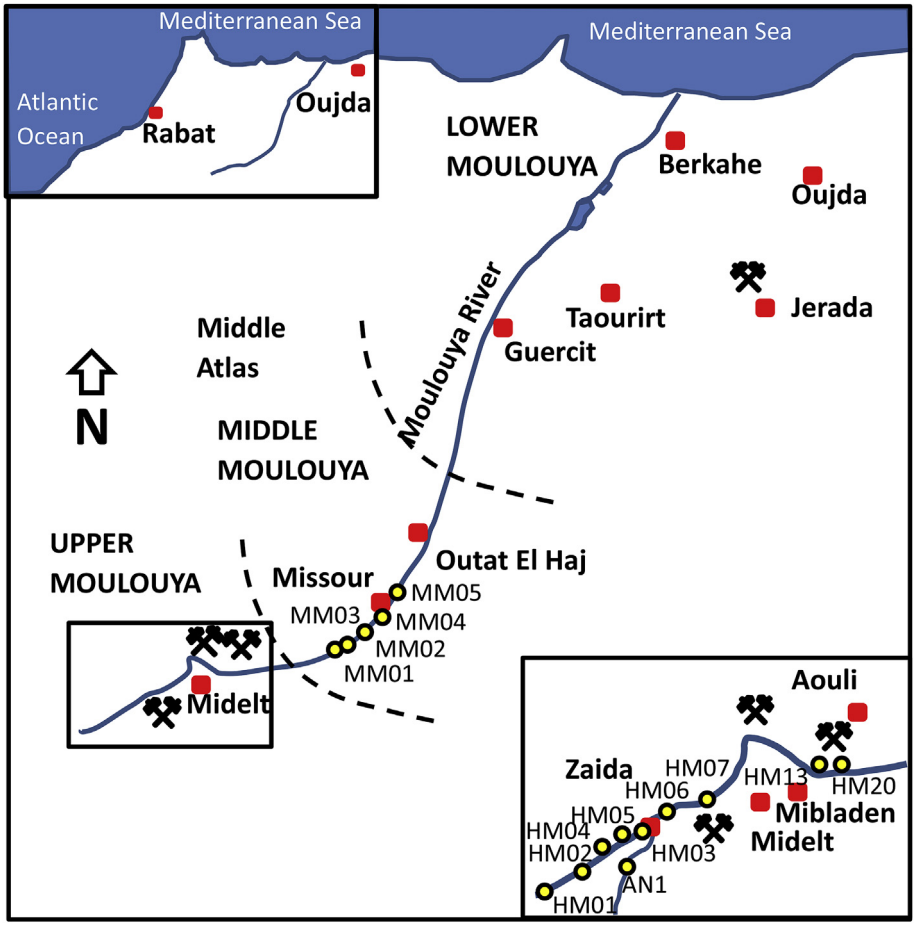

Fig. 1. Map of study area showing the Moulouya river and the sampling points of this work.

presented by Iavazzo, Adamo, Boni, Hillier, and Zampella (2012).

In the Upper Moulouya Lead district there are three main lead deposits: Zaida, Mibladen and Aouli (Fig. 1) that were exploited from 1972 to 1985, 1936 to 1985 and 1926 to 1985, respectively (Iavazzo et al., 2012). Mining activities have created abandoned mine sites comprised of: waste deposits, pit lakes, processing plants and waste tailings. Leaching and erosion are processes, among others, that can introduce metals and natural radionuclides into the Moulouya River and its tributaries where heavy metals and natural radionuclides could increase.

The heavy metal contamination of the Moulouya river from the abandoned mines of Aouli, Mibladen and Zaida was studied by Makhoukh, Sbaa, Berrahou, and Vanclooster (2011), who observed high concentrations of metals in the vicinity of abandoned mines and facilities. However, no similar studies concerning NORM have been performed in this ecosystem.

A sampling campaign was performed in 2010. Water was collected directly from the Moulouya river, some tributaries and three pit lakes. These waters were transferred into 25 L plastic bottles and 10 ml of HNO_3 were added to avoid adsorption by plastic walls. Sediment samples were collected directly at the same points as the water and stored in plastic bags.

The sampling stations are shown in Fig. 1 and some physico-chemical parameters of the waters are compiled in Table 1. The pH of the water samples shows slight alkalinity due to the presence of carbonates (Makhoukh et al., 2011). Higher pH values (around pH = 9) were also measured in the pit lake waters where the mineral is also carbonated (Makhoukh et al., 2011; Iavazzo et al., 2012) and where the nature of sediments would also be affected by mining. The high pH values are in agreement with the alkalinity of the pit lake sediments and the river substrates. The high percentage of CaO (36.77%) in the sediments of this river has been shown to be is related to its limestone nature (Bounakhla et al., 2012).

Table 1
Values of pH, conductivity and TDS in waters.

Sample	pH	Conductivity (mS/cm)	TDS g/l	Notes
Upper Moulouya				
HM01	8.14	3.5	1.9	Not contaminated
HM02	7.56	3.6	1.9	Urban wastes
AN1	8.28	3.7	2	Tributary
HM03	8.07	3.6	1.9	Zaida
HM06	8.09	3.6	1.9	Zaida
HM13	7.86	3.3	1.8	Aouli
HM20	8.58	3.3	1.8	Aouli
Middle Moulouya				
MM01	7.86	4.8	2.6	
MM02	8.24	4.5	2.4	
MM03	8.15	5.1	2.7	
MM04	8.21	5.1	2.7	
MM05	8.06	5.5	2.9	
Pit lakes				
HM04	9.15	3.5	1.9	Pit lake
HM05	9.57	3.6	1.9	Pit lake
HM07	9.23	3.4	1.8	Pit lake

The conductivity in the Upper Moulouya river is in the range of 3.3–3.7 mS/cm for the river water samples and 3.4–3.6 mS/cm for the pit lake waters. These high values can be explained by the high mineralization of the waters (Makhoukh et al., 2011). Conductivity is higher in samples collected from the Middle Moulouya river (4.8–5.5 mS/cm), because these waters could be affected by a dam

located upstream (between HM20 and MM01) and by urban waste.

For radionuclide determination in the waters, an aliquot of 1 L which was previously acidified was taken and initially an accurate activity of ^{232}U, ^{229}Th and ^{209}Po was added for radiochemical yield determination. Afterwards the actinides and polonium were co-precipitated from the waters with $Fe(OH)_3$ (Holm & Fukai, 1977) and the precipitate was separated from the aqueous solution by centrifugation. After drying, the precipitate was dissolved with 10 ml of 8 M HNO_3.

The sediments were dried, powdered and sieved after their collection and representative aliquots were traced with an accurate activity of ^{232}U, ^{229}Th and ^{209}Po and dissolved with acid (Lee & Lee, 2001) for radionuclide determination. The dissolution was performed using first aqua regia with H_2O_2 and afterwards nitric acid 60%, being the obtained solution evaporated until it is dry and then dissolved with 8 M HNO_3. From this solution, actinides were co-precipitated with Fe^{3+}, and the precipitate was separated from the solution by centrifugation, dried and finally dissolved in 10 ml of 8 M HNO_3.

From the obtained 10 ml 8 M HNO_3 solutions (in both waters and sediments) the sequential isolation of the Uranium-isotopes, the Thorium-isotopes, and ^{210}Po was performed by using UTEVA (Uranium and TEtraValents Actinides, Eichrom trade mark) chromatographic resins. The radiochemical procedure, based on the use of UTEVA chromatographic resin as described by Casacuberta et al. (2012), provides three aqueous solutions containing, in an isolated form, the U-isotopes, the Th-isotopes and Po-210.

The Uranium and Thorium sources for alpha-particle spectrometric measurements were prepared by the electrodeposition of the elements onto stainless steel planchets following the method of Hallstadius (1984). With this purpose the aqueous solutions containing Uranium or Thorium after application of the selected radiochemical procedure are evaporated until they are near to being completely dry and conditioned as described by Mantero, Lehritane, Hurtado, and García-Tenorio (2010). The electrodeposition arrangement depends on the radionuclide of interest, using the electrical intensity 1.2 A in the case of Uranium and 1.5 A for Thorium.

On the other hand, a well-established method (Flynn, 1968) based on the self-deposition of Polonium on a silver or copper disc is used for obtaining radioactive sources of ^{210}Po from the aqueous solutions containing this radionuclide. The adaptation of the Flynn method to our laboratory conditions is described in detail by Díaz-Francés, Mantero, Manjón, Díaz, and García-Tenorio (2013).

Uranium-isotopes (^{238}U, ^{235}U, ^{234}U), Thorium-isotopes (^{230}Th, ^{232}Th) and ^{210}Po activity determinations were performed by applying the alpha-particle spectrometric technique using an Alpha-Analyst, spectrometer manufactured by Canberra Co. The system is formed by eight independent chambers, each chamber being equipped with a 450 mm^2 PIPS detector, model A450-18AM, manufactured by Canberra Co. Only two chambers were used for ^{210}Po measurements, two other chambers were used for Thorium-isotopes measurements and the remaining four chambers were used for Uranium-isotopes determinations. Activity determinations were performed by applying the isotope-dilution technique.

3. Results and discussion

The U-isotopes and ^{210}Po results obtained in the analysis of the water samples are listed in Table 2. ^{238}U activity concentrations range from 9.4 to 203 mBq/l (Upper Moulouya river), from 6.6 to 22.8 mBq/l (Middle Moulouya River), and from 235 to 1979 mBq/l (pit lakes of abandoned mines), while the $^{234}U/^{238}U$ activity ratios range from 1.9 to 3.4 (Upper Moulouya river), from 1.8 to 2.0 (Middle Moulouya river), and 4.3–5.4 in the pit lake water samples. The ^{210}Po activity concentrations in the same waters were in the range of 0.7–2.7 mBq/l (Upper Moulouya River), 1.5–13.2 mBq/l (Middle Moulouya river), and 2.3–4.7 mBq/l (pit lake water samples).

Mining has previously been considered a source of contamination in the Moulouya river and pit lakes, in the zone of Zaida, when trace metals were only considered ((Bounakhla et al., 2012). The results obtained in this work indicate that this last conclusion can be extended to include U-isotopes. The evidence that mining activities can be a potential source of uranium in to the nearby environment is reflected in the higher activity concentrations found for ^{238}U and ^{234}U in the waters of the Moulouya river located near Zaida (a mining area), which reached one order of magnitude higher than typical levels of uncontaminated rivers (Martínez-Aguirre & García-León, 1992) (Table 2). Additionally, the U-isotope activity concentrations in the three pit lakes are one to two orders of magnitude higher than in uncontaminated waters collected in rivers of southern Spain (the high levels of uranium in dissolution in the pit lakes is an indication of the potential tendency of this radionuclide to leach from wastes, tailings and residues located in the surroundings that can finish in the river).

Table 2
U-isotopes and ^{210}Po activity concentrations (mBq/l) in water samples.

Sample Code	^{238}U	^{234}U	$^{234}U/^{238}U$ activity ratio	^{210}Po
Upper Moulouya: river water samples				
HM01	9.4 ± 0.5	21.4 ± 0.8	2.28 ± 0.15	0.9 ± 0.2
HM02	17.9 ± 1.1	34.3 ± 1.6	1.92 ± 0.15	2.0 ± 0.3
AN1	3.6 ± 0.4	7.6 ± 0.6	2.13 ± 0.29	2.7 ± 0.3
HM03	177 ± 5	57 ± 13	3.25 ± 0.11	1.1 ± 0.2
HM06	203 ± 4	702 ± 12	3.45 ± 0.09	2.5 ± 0.3
HM13	56 ± 2	117 ± 3	2.07 ± 0.09	2.3 ± 0.3
HM20	14 ± 1	27.6 ± 1.4	1.92 ± 0.16	0.7 ± 0.1
Middle Moulouya: river water samples				
MM01	21 ± 1	38.2 ± 1.4	1.80 ± 0.11	4.0 ± 0.4
MM02	10.9 ± 0.7	20.1 ± 1.0	1.85 ± 0.15	13.2 ± 0.8
MM03	6.6 ± 0.6	10.5 ± 0.8	1.60 ± 0.18	12.0 ± 0.8
MM04	14.7 ± 0.9	28.5 ± 1.3	1.94 ± 0.14	3.8 ± 0.5
MM05	22.8 ± 0.9	46.1 ± 1.5	2.02 ± 0.10	1.5 ± 0.2
Pit lake water samples				
HM04-3	1979 ± 169	8564 ± 671	4.33 ± 0.50	2.7 ± 0.3
HM05-3	443 ± 8	1921 ± 32	4.34 ± 0.11	2.3 ± 0.3
HM07	235 ± 14	1267 ± 62	5.40 ± 0.41	4.7 ± 0.5
Typical uncontaminated level range				
Uncontaminated	11–32 [a]	13–40 [a]	1.0–1.6 [a]	3 [b]

[a] Martínez-Aguirre & García-León, 1992.
[b] Persson & Holm, 2011.

In summary, mining can be considered as a U source that is impacting on the Moulouya aquatic system as this can be deduced from the results obtained in sampling points HM03 and HM06 located in Zaida City, in the vicinity of abandoned mines, where the ^{238}U activity concentrations reach 203 ± 4 mBq/l. Similar conclusions can be deduced from the analysis of the ^{238}U results corresponding to the sampling points located in the Aouli mines area (particularly HM13).

However, according to Table 1, pH and conductivity have similar values in all the samples analyzed and a relationship between U-isotope activity concentration to pH or conductivity is not observed. The high radionuclide levels can therefore only be related to former mining activities.

The presence of U in dissolution is favored by the formation of soluble uranium carbonated complexes when there is a leaching process from the treated substratum. The presence of Uranium dissolved in water can thus be explained by the formation of carbonates (Broecker, 1974).

The existence of U impact in the Upper Moulouya river is reinforced by observing the obtained $^{234}U/^{238}U$ activity ratios in the waters analyzed. In the river samples with higher Uranium concentrations, such as HM06 the value of this activity ratio (3.45 ± 0.09) is in good agreement with the $^{234}U/^{238}U$ activity ratios found in the pit lake waters which were in the range of 4.3–5.4. $^{234}U/^{238}U$ activity ratios higher than unity are usually explained by the nucleus recoil after alpha particle emission. In a lixiviation process ^{238}U daughters (^{234}Th, ^{234}Pa and ^{234}U) would have enough energy to break the chemical bonds (Coward

Table 3
^{232}Th, ^{230}Th and ^{234}U activity concentrations (mBq/l) in waters.

Sample Code	^{232}Th	^{230}Th	^{234}U
River water samples			
HM01	0.5 ± 0.2	1.7 ± 0.3	21.4 ± 0.8
HM02	0.4 ± 0.2	1.1 ± 0.3	34.3 ± 1.6
HM03	0.8 ± 0.4	1.6 ± 0.5	7.6 ± 0.6
AN1	0.8 ± 0.3	2.9 ± 0.6	576 ± 13
HM06	0.32 ± 0.14	2.4 ± 0.4	702 ± 12
HM13	1.3 ± 0.5	8.4 ± 1.1	117 ± 3
HM20	1.1 ± 0.3	4.2 ± 0.7	27.6 ± 1.4
MM01	1.4 ± 0.3	3.5 ± 0.5	38.2 ± 1.4
MM02	3.0 ± 0.6	7.0 ± 0.9	20 ± 1
MM03	0.12 ± 0.09	0.1 ± 0.05	10.5 ± 0.8
MM04	0.2 ± 0.1	1.2 ± 0.3	28.5 ± 1.3
MM05	0.9 ± 0.3	2.2 ± 0.4	46.1 ± 1.5
Pit lakes water			
HM04	0.2 ± 0.1	0.7 ± 0.2	8564 ± 671
HM05	< AMD ±	2.8 ± 0.2	1921 ± 32

Table 5
^{232}Th, ^{230}Th and ^{234}U activity concentrations (Bq/kg) in sediments.

Sample Code	^{232}Th	^{230}Th	^{234}U
River sediment samples			
HM01	14.9 ± 0.9	9.6 ± 0.7	4.9 ± 0.9
HM02	24.5 ± 1.7	14.3 ± 1.2	8.8 ± 1.0
AN1	11.7 ± 1.1	8.5 ± 0.9	12.9 ± 0.7
HM03	18.4 ± 1.2	11.5 ± 0.9	197 ± 7
HM06	13.7 ± 1.2	10.6 ± 1.0	23.4 ± 1.4
HM13	14.9 ± 0.9	15.0 ± 0.9	13.0 ± 0.8
HM20	23.7 ± 1.6	16.2 ± 1.0	13.3 ± 0.7
MM01	12.7 ± 0.8	15.3 ± 0.9	11.0 ± 0.6
MM02	9.6 ± 0.7	14.3 ± 0.9	11.2 ± 0.6
MM03	13.4 ± 0.7	16.6 ± 0.8	12.3 ± 0.7
MM04	15.8 ± 0.8	17.8 ± 0.9	12.6 ± 0.6
MM05	17.1 ± 0.9	17.5 ± 0.9	13.9 ± 0.8
Pit lakes sediment samples			
HM04	13.7 ± 1.0	38.8 ± 1.9	97 ± 3
HM05	14.9 ± 0.9	26.2 ± 1.2	44.5 ± 1.4

& Osmond, 1974).

In the case of ^{210}Po, the activity concentrations found along the entire water course were similar to the ones found in uncontaminated rivers (Persson & Holm, 2011). Quite low activity levels are found in the pit lakes waters and in all the Moulouya river waters regardless of the place of sampling collection. Undetectable enhanced levels were observed in the upper Moulouya river waters collected in the vicinity of the former mining activities. This lack of ^{210}Po environmental impact is not surprising due to the tendency of this radionuclide to be present in aquatic systems associated with particulate matter and its very low tendency to leach (Barbero et al., 2014).

In all the waters the activity concentration of Th-isotopes (^{232}Th and ^{230}Th) was also determined and the results are shown in Table 3. The activity concentrations of ^{232}Th found were to be in the range of 0.2–1.3 mBq/l, while the activity concentrations of ^{230}Th were from 0.1 to 8.4 mBq/l across the whole set of water samples. This set of results reflects a lack of this element's impact in the Moulouya river. Similarly to Po, the tendency of Th is to remain associated to solid fractions in aquatic systems. The distribution coefficient of this element, in surface waters, is usually one or two orders of magnitude higher than those observed for U. Even in carbonated solutions the Th did not form soluble compounds. In surface aquatic systems the ^{230}Th/^{234}U activity ratio is usually very low (Ku, 1976).

The ^{238}U, ^{234}U, ^{210}Po, ^{230}Th and ^{232}Th activity concentrations determined in sediment samples are listed in Tables 4 and 5. The U levels found were, in general, quite moderate (with the exception of the HM03 sample) with ^{238}U activity concentrations ranging from 4.0 to 15.2 mBq/g in Upper Moulouya, from 8.8 to 11.9 mBq/g in Middle Moulouya and from 28 to 39 mBq/g in pit lakes. The corresponding ^{234}U/^{238}U activity ratios were in the range of 1.1–2.2 in sediments collected along the whole of Moulouya river, while this activity ratio ranges from 2.5 to 3.8 in the pit lake sediments. The U impact observed in the waters is not unambiguously distinguishable in the sediments, which is in agreement with the hypothesis that the U incorporated in to the Moulouya river is mostly dissolved after leaching in the abandoned mining area and afterwards is transported to the river's watercourse where is diluted.

Table 4
U-isotopes and ^{210}Po activity concentrations (Bq/kg) in sediment samples.

Sample Code	^{238}U	^{234}U	^{234}U/ ^{238}U activity ratio	^{210}Po
Upper Moulouya: river sediment samples				
HM01	4.0 ± 0.8	4.92 ± 0.9	1.46 ± 0.14	43.3 ± 2.3
HM02	7.9 ± 0.9	8.80 ± 1	1.2 ± 0.3	71 ± 8
AN1	7.6 ± 0.6	12.93 ± 0.8	1.7 ± 0.2	28.0 ± 1.2
HM03	52 ± 2	197.00 ± 7	1.1 ± 0.2	1.7 ± 0.5
HM06	8.8 ± 0.8	23.40 ± 1.4	2.16 ± 0.14	31 ± 5
HM13	9.8 ± 0.8	13.00 ± 0.8	1.2 ± 0.1	
HM13F	11.1 ± 0.6	13.29 ± 0.7	1.20 ± 0.09	54 ± 2
HM20	15.2 ± 0.8	17.64 ± 0.9	1.16 ± 0.09	39.4 ± 1.4
Middle Moulouya: river sediment samples				
MM01	9.3 ± 0.6	11.04 ± 0.6	1.2 ± 0.1	40 ± 2
MM02	8.8 ± 0.5	11.22 ± 0.6	1.3 ± 0.1	46 ± 2
MM03	10.1 ± 0.6	12.27 ± 0.7	1.2 ± 0.1	52 ± 2
MM04	10.5 ± 0.6	12.63 ± 0.6	1.20 ± 0.09	72.5 ± 2.5
MM05	11.9 ± 0.7	13.87 ± 0.8	1.17 ± 0.09	29.0 ± 1.2
Pit lake sediment samples				
HM04	38.5 ± 1.7	97 ± 3	3.8 ± 0.2	62 ± 8
HM05	28 ± 1	44.5 ± 1.4	2.52 ± 0.14	
HM07				16 ± 4

The activity concentrations of ^{210}Po in the same sediments were in the range of 31–71 mBq/g along the Upper Moulouya river, 28–72 mBq/g along the Middle Moulouya river and 16–62 mBq/g in the pit lake sediments. In the case of ^{230}Th the activity concentrations along the Moulouya river were between 8.5 and 17.5 mBq/g, whereas the range in the pit lake samples was 26–39 mBq/g. For both elements, Th and Po, the activity levels found are in the range of values observed in riverbed sediments worldwide which are not affected by anthropogenic inputs (Gonzalez-Labajo, Bolívar, & García-Tenorio, 2001).

Some additional information about the behavior of the natural radionuclides analyzed and the possible anthropogenic impact generated by the former mining activities in the Upper Moulouya catchment area can be obtained through the study of the distribution coefficients (K_d) found for the analyzed radionuclides along the whole Moulouya watercourse. The determinations performed at the same sampling point in both waters and sediments enables the determination of the distribution coefficients (K_d, l/kg), defined as the quotient of ^{238}U activity concentration in sediment (mBq/kg) and water (mBq/l). In Fig. 2 the K_d values for U and Po are shown for comparison, which led to the following comments:

a) The K_d obtained for Po-210, as expected, is clearly higher than the K_d of Uranium due to the known tendency of the Po to be bound to particulate matter in opposition to the more conservative behavior of Uranium in aquatic systems.
b) A good correlation is found between the K_d(U) values and the Uranium concentrations determined in the waters, as shown in Fig. 3. K_d decreases when the Uranium concentrations in the waters increase. Higher K_d(U) values, reaching 10^3, were determined in the sampling points less influenced by mining, while values as low as 10^1 were determined in the pit lakes, indicating the tendency of the uranium to be in dissolved form in environments rich in carbonates. The sediments in the pit-lakes show higher U activity concentration values than those in the riverbed sediments indicating that a fraction of anthropogenic U can accumulate, but it has a higher tendency to

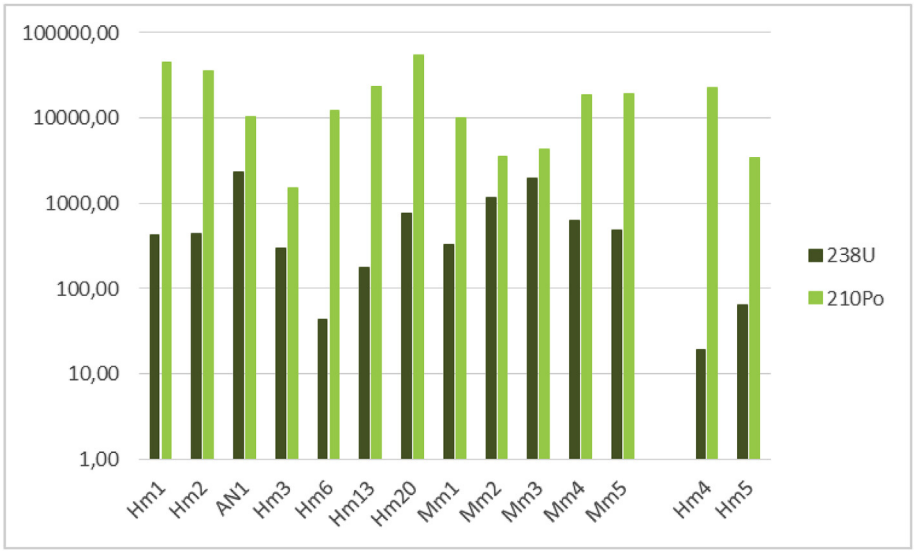

Fig. 2. Distribution coefficients (K_d, l/kg) for U and Po, defined as the quotient of activity concentration in sediment (mBq/kg) and water (mBq/l).

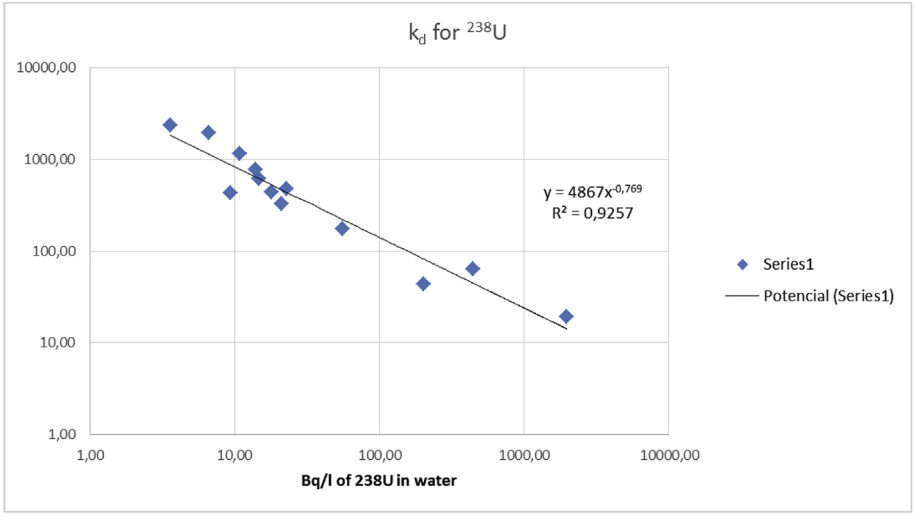

Fig. 3. K_d(U) versus ^{238}U activity concentrations in the water samples.

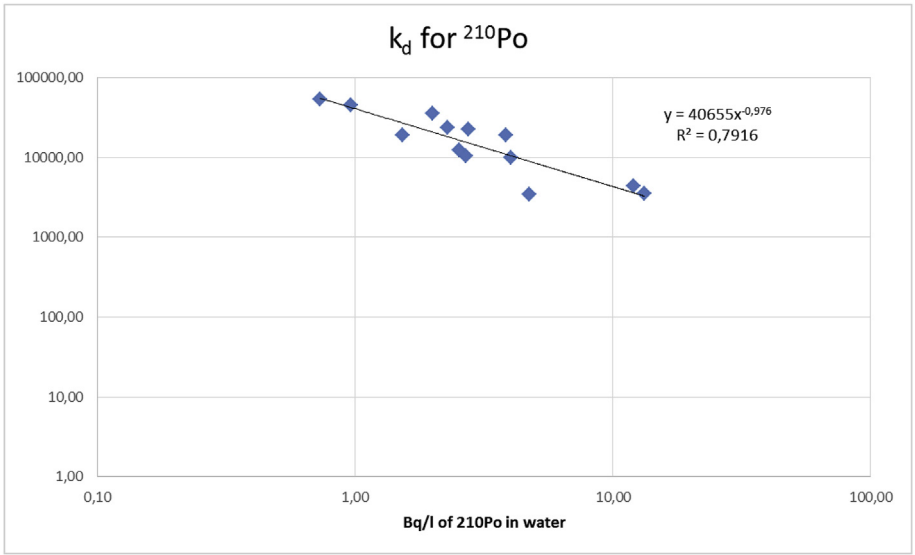

Fig. 4. K_d(Po) versus ^{210}Po activity concentrations in the water samples.

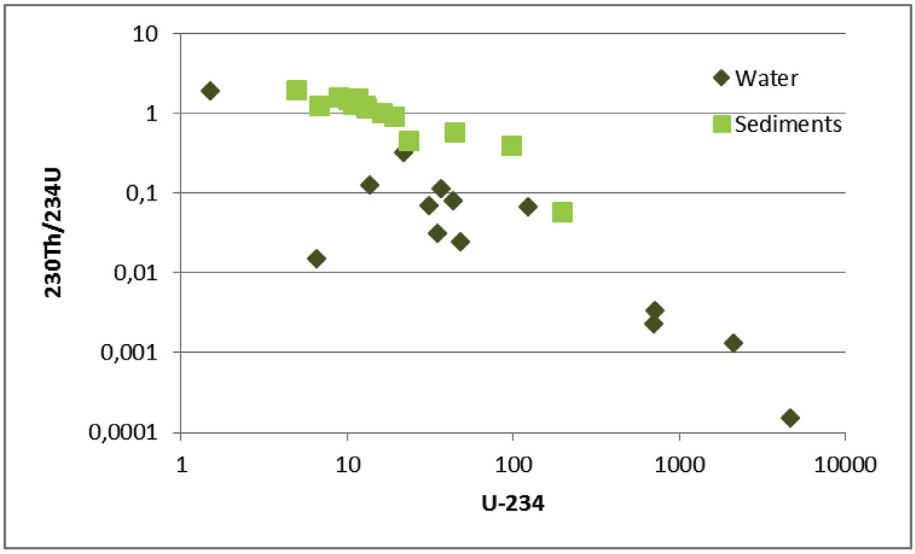

Fig. 5. $^{230}Th/^{238}U$ versus ^{234}U activity concentrations activity ratios in waters and sediments.

dissolve. Consequently, the sediments of the analyzed pit lakes cannot be considered a sink of U, in the sense that the majority of the U existing in the water ecosystem does not accumulate.

c) The K_d of Po is less sensitive to the concentrations found for this radionuclide in the waters, as can be observed in Fig. 4.

These conclusions reinforce previous comments which state that nowadays the impact of former mining activities in the Moulouya river waters is restricted only to the presence of Uranium, due to its tendency to be in dissolved form as carbonates, while no influence is observed for ^{210}Po and Th isotopes.

The analysis of the $^{230}Th/^{234}U$ activity ratios also reinforces the main conclusion of this work. This activity ratio has a value near unity in the majority of the samples (waters and sediments), particularly those from sampling points located far from the mining area (Fig. 5). However, the ratio is lower than 1 in the sediment pit lakes and in the sampling points near the abandoned mines, again indicating the presence of some anthropogenic Uranium. In the case of the river waters, the $^{230}Th/^{234}U$ activity ratio was in the range of 10^{-2} to 1 with a tendency for activities ratios to decrease when the Uranium concentration in water increases. This activity ratio is in the range of 10^{-4} to 10^{-2} in the water samples collected in pit lakes and in a river water sample (HM06) located not far from the mines, reflecting the tendency of Uranium to be in dissolved form.

4. Conclusions

Natural radionuclides (U-isotopes, ^{210}Po, ^{230}Th and ^{232}Th) activity concentrations were measured in samples of water and sediment collected along the Moulouya river (east Morocco) and in some pit lakes formed in abandoned mines nearby. High Uranium concentrations were found in the waters of pit lakes formed by former open air mines; these levels are associated with the presence of carbonates. In addition, relatively high levels of U were also observed in water taken from sampling points of the Moulouya river located in the vicinity of former mines, indicating the local impact of the former mining activities on the aquatic system. The impact of U in the waters of the Upper Moulouya region was confirmed through the analysis of the corresponding K_d(U) values and the $^{230}Th/^{234}U$ activity ratios. Although the mining impact is not easily determined when U levels in the sediments of the Moulouya river are compared, the lower values of the $^{230}Th/^{234}U$ determined in the sediments collected in the vicinities of the abandoned mines could be considered as an impact evidence. However, non-enhanced levels in waters and sediments were found for ^{210}Po, ^{230}Th and ^{232}Th, reflecting the lower tendency of these radionuclides to be in dissolution even in solutions rich in carbonates. The impact of the former mining activities in relation to these last radionuclides should be considered as negligible.

Conflicts of interest

None declared.

Ethical statement

Authors state that the research was conducted according to ethical standards.

Funding

This work has been partially financed by the Spanish Agency of International Cooperation and Development (AECID) of the Government of Spain.

Acknowledgments

This work has been partially financed by the Spanish Agency of International Cooperation and Development (AECID) of the Government of Spain. The authors gratefully acknowledge the laboratory assistance of J. Díaz.

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