

# Assessment of Natural Radioactivity (Uranium, Thorium and Potassium) and heavy metals Contents in Water and Sediments from North Africa dams (Algeria and Tunisia): Geochemical behavior and environmental implications

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## Article

# Impact of Natural Radioactivity (Uranium, Thorium and Potassium) and Toxic Heavy Metals Contents in Water and Sediments from North Africa Dams (Algeria and Tunisia): Geochemical Behavior and Environmental Implications

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**Abstract:** Natural radioactivity of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  for liquid and solid samples collected from Sidi Salem dam (Tunisia) and Aïn Dalia dam (Algeria) were measured using TERRA detector of rays (gamma, beta, and alpha), Atomic adsorption and Gamma-ray spectrometry were used to analyze the levels of radionuclides and toxic heavy metals, respectively. Toxic heavy metals (Fe, Bb, Zn, Ni, Cu, Cr and Cd) and associated health risks in surface water and sediment of dams have been investigated in this present study. The radioactivity mean rates in the water samples were  $1.72 \pm 0.01$ ,  $0.068 \pm 0.01$  and  $94.6 \pm 1.04$  BqL<sup>-1</sup> for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively (Tunisia dam) and were  $1.9 \pm 0.24$ ,  $0.09 \pm 0.01$  and  $131.43 \pm 1.03$  BqL<sup>-1</sup> for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively (Algeria dam). The  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radioactivity mean concentration measured in the sediment samples were  $2.67 \pm 0.01$ ,  $0.18 \pm 0.012$  and  $197.87 \pm 2.01$  Bqkg<sup>-1</sup> respectively (Tunisian dam) and were  $4.34 \pm 0.05$ ,  $0.27 \pm 0.05$  and  $287.61 \pm 3.34$  Bqkg<sup>-1</sup> respectively (Algeria dam). The activity concentration of  $^{40}\text{K}$  was higher than that of  $^{238}\text{U}$  and  $^{232}\text{Th}$  for the water and sediment samples. The mean activity concentrations follow the order  $^{40}\text{K} > ^{238}\text{U} > ^{232}\text{Th}$ . The cumulative impact can be a serious immediate radiological health burden on the ecosystem, environment and people's health in this study area and can be transposable to any other similar region. The good knowledge of monitoring quality and quantity for the transboundary water resources and the international collaborations are essential to safeguard human health (women's breasts cancer, thyroid cancer, neurological impact...), and avoid the conflicts, especially during these climatic upheavals of drought.

**Keywords:** Natural radioactivity; Water; Sediment; environmental implications; North Africa

## 1. Introduction

Natural radiation is a normal part of the environment that emanates from two main sources: cosmic radiation, which originates in outer space and passes through the atmosphere, and the decay of radionuclides (radioactive isotopes or radio-isotopes) in the soil and the rock. Radionuclides undergo spontaneous disintegration into daughter nuclides with an associated emission of ionizing radiation in the form of alpha and beta particles, gamma rays and other cosmic rays. Daughter nuclides may be either stable or may themselves be radionuclides which also undergo radioactive decay. Many people might be surprised to learn that drinking water sources, especially rainwater,

surface water and groundwater, can contain radioactive elements (radionuclides). Radionuclides in water can be a concern for human health because several are toxic or carcinogenic. Other radionuclides are useful tools for determining the hydrodynamic of groundwater, the residence time, the radioactivity rate disintegration, the water quality, the contamination source/rate, the age of groundwater in an aquifer or of sediment deposited at the bottom of a water body (dead zone of the dams).

The Majerda transboundary basin, a prominent watershed in North Africa, extends from Eastern Algeria, near Souk Ahras, and stretches into Tunisia. Covering a vast expanse of approximately 23,175 square kilometers, with around 7,700 square kilometers lying within Algeria's borders, it plays a crucial role in the region's water resources. The exploitation of Uranium-rich ore deposits such as diapiric rocks in the Atlas of Algeria (Souk Ahras basin), together with nuclear development around the world, has increased the risk of anthropogenic radioactive pollution. Protection against such risks is essential. Aware of the importance of the problem, the Tunisian government has already launched a program that envisages the development of studies of the radiological environment in collaboration with the International Atomic Energy Agency (IAEA) and with the World Health Organization (WHO).

This study was initiated in the transboundary basin (Tuniso-Algerian basin) known, in particular, by its richness in mines (phosphate, Pb/Zn, Fe...), diapiric sediments (clay, limestone, gypsum, salt...) and volcanic (magmatic and metamorphic rocks) deposits characterized by an average Uranium content of 35 ppm. The main objective of this study is to make in situ measurements of the natural gamma radioactivity and the geochemical analysis of ( $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ ) and the toxic elements in the greatest dam in Tunisia (Sidi Salem dam) and Aïn Dalia dam from Algeria (Tuniso-Algerian transboundary basin) and to estimate the naturally occurring radioactivity in the surface water and the sediment from the dead zone of the dams.

The natural radioactivity radiation comes in many forms (varieties of cosmic rays) [1]. Human health exposure to a high concentration of background radiation (cumulative effect) may cause health risks. The taking radionuclides naturally present in the atmospheric environment, water resources (surface and groundwater), sediment or food above the international limit can cause cardiovascular epidemic and other effects of excessive radiation exposure dosing during the time [1]. In this current study area, water quality deterioration has been reported and the dependence of the inhabitants on the water supply available without security guarantees [2, 3]. Many scientific reports have been conducted on the direct investigation of radioactivity levels in soil, water, food and sediment around the world, including North African countries (Tunisia and Algeria) [4, 5]. No immediate harmful effects on health due to the exposure of diapiric/metamorphic rocks but in the long term by cumulative effect everything is possible and plausible. Several countries in the world suffer from this type of problem such as Tunisia, Algeria, Saudi Arabia, Kuwait, Iran, Qatar, Oman, Spain, Portugal, Niger, and Nigeria. In southern Tunisia, recent studies carried out by Hamed et al. [6], have shown high rate of radioactivity (U, Th, Rd...) and toxic heavy metals (Cd, Pb, Zn, Fe, Cr, Sr, Li...), which have exceeded the standards in the phosphate/petroleum and geothermal basin (North Western Sahara Aquifer System: NWSAS-the great transboundary basin of North Africa, divided between Algeria, Tunisia and Libya). The damage to human health is visible by an abnormal level of cancer disease in this mining polluting basin (Gafsa, Sfax, Gabes, Tozeur and Kebili), which affects all ages and both sexes. Therefore, the aim of this scientific report is to assess the radiological impact and deduce the level of possible risk from the level of radioactivity and toxic heavy metals measured in surface water and sediments in the Aïn Dalia dam in the Algerian territory (recharge zone) and the same thing in the biggest dam of Sidi Salem in Tunisian territory (discharge area).

## 2. Study area

This transboundary basin is located in North Africa between Algeria (recharge area) and Tunisia (discharge area) Southern of the Mediterranean Sea (Figure 1). This basin was characterized by specific geomorphology influenced by the Euro-African collision plates. It is characterized by the existence of natural volcanic deposits (diapiric, magmatic, and metamorphic rocks) in the North of

the Atlas transboundary Tuniso-Algerian basin. The hydrographic network is dendritic type, it is made up of a single permanent watercourse (Majerda River) belonging to the cross-border Majerda-Mallegue watershed, the upstream of which is in the Algerian territory and the downstream in the Tunisian territory. The entire hydrographic network converges towards the Tunisian Mediterranean coast/*Carthage Sea*.

The study area is famous for its huge number of dams in both territories and iron, Pb, and Zn ore deposits and industrial sectors from these mine minerals...which were part of the raw materials of the large ironwork in North Africa and cement works. The Majerda basin falls within the sub-humid climatic zone, characterized by very high rainfall (500 - 1200 mm/y) and dry seasons and it is located in the trough of the great collapse ditch of the Majerda [7]. The study area is characterized by a superposition of multi-layer aquiclude/aquitard [8] with the exception of the perched karstic carbonate (limestone and dolomite) units in NE Algeria and in NW Tunisia which are very vulnerable to this type of pollution by toxic and radioactive elements, or the possibility of contamination is possible by fracturations and/surface and subsurface faults (decreasing the residence time). There are therefore significant surface reserves that are used in agricultural, domestic, industrial, and tourist areas.

The Aïn Dalia dam is located in the town of Annaba in the Majerda river (Algerian part), their coordinates are latitude 36.263439°N and longitude 7.862016°E. It has been built since 1956 to store surface water and protect cities against flooding. Concerning the geographical locations, the Sidi Salem dam is located in the town of Beja (Testour) in the Majerda River (Tunisian part). The coordinates position are latitude 36.590614°N and longitude 9.396958°E. The dam has been built since 1977 to also store surface water and use it during periods of shortage. The dimension of the foot of this ancient gigantic dam is 73 meters in height and 345 meters in length. The principal objectives of this dam are the artificial recharge of aquifers, use in tourism, and industrial activities in the coastal part of Tunisia by pipeline transfer (Bizerte, Tunis, Hammamet, Sousse, Monastir, Sfax...).

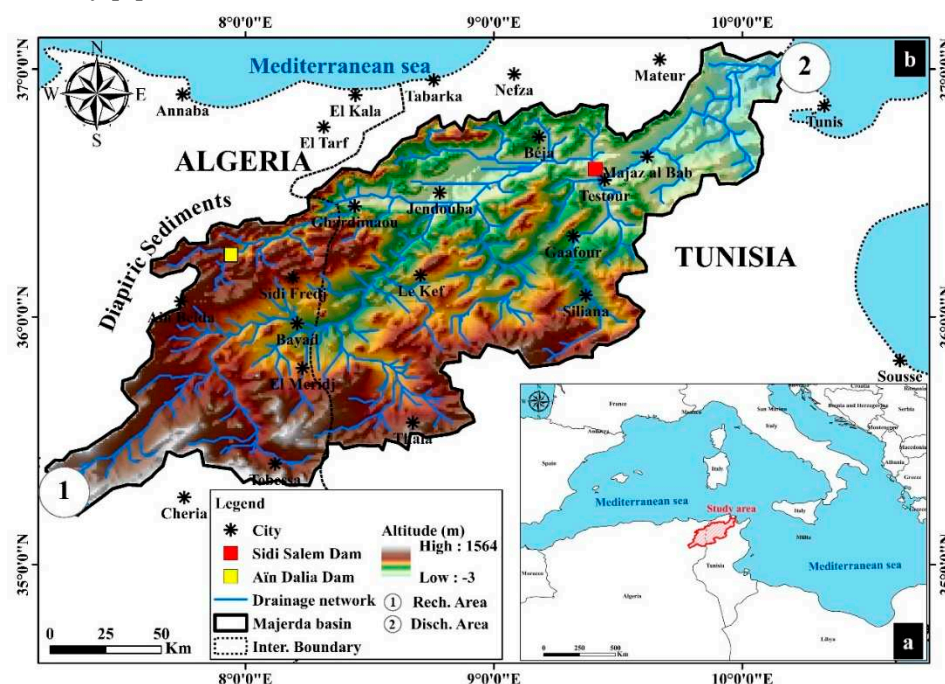


Figure 1. Geographic location of the transboundary basin of the study area. .

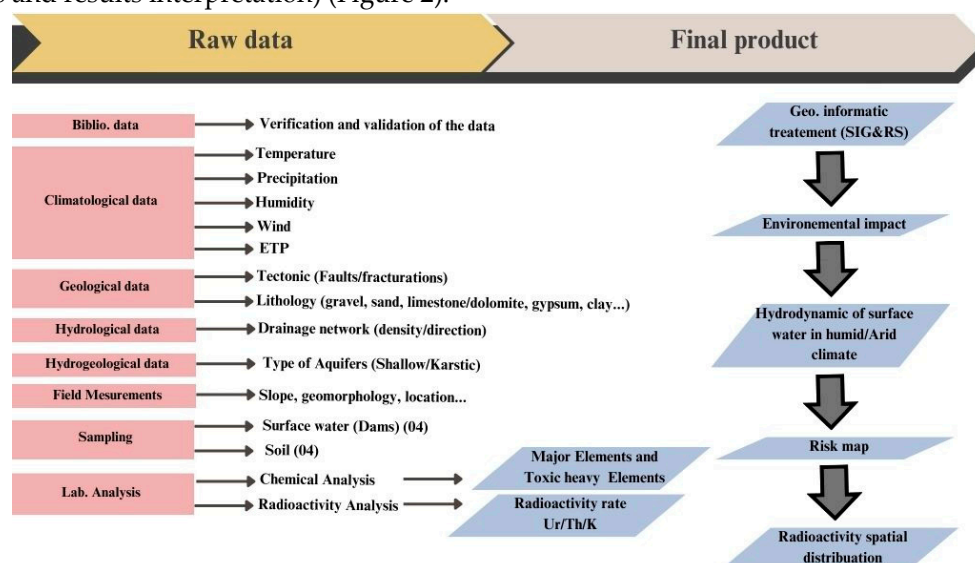
### 3. Materials and methods

#### 3.1. Work methodology

Our study consists of following international steps and precautions. A combination of several factors was carried out during this study. The flowchart in the following figure summarizes all the



steps (Geological/climatological data, field measurements, sampling water/sediment, laboratory analysis and results interpretation) (Figure 2).



**Figure 2.** Flowchart of the work methodology in the study area.

### 3.2. Sample collection and preparation

In this present research, 08 samples (04 surface water and 04 solids) have been collected during spring 2023, liquid and sediment parts of the 02 dams (Aïn Dalia and Sidi Salem). The water container is sampled and washed with chloric acid (acidified 0.1 M HCl), at a rate of 10 ml per liter to avoid any kind of contamination according to international protocol. The sediment samples were placed directly in transparent cellophane bags to avoid any kind of evaporation and contact with open air (avoid the contact air/sediment). 1.5 liters of liquid water and 2.5 kg of sediment were collected, labeled and properly sealed. All the bottles without exception were first washed according to international protocol with sulfuric acid (diluted H<sub>2</sub>SO<sub>4</sub>) then rinsed with distilled water and then with dam water to avoid any external physical and/or biological contamination (temperature, humidity, and pressure).

### 3.3. Gamma Counting

Terra detector (Ukraine-made / April-2021) was used for the gamma counting at the Faculties of Sciences of Sciences of Gafsa (Tunisia) and the Geolab (Sétif-Algeria). The chemical analysis has been effected in the Sfax Tina/lab (Tunisia). The radioactivity detector has a resolution of 8%. A detailed description of isotopic disintegration counting has been reported by numerous specialists in the field of radioactivity [12, 13]. The radioactive activity concentrations in the samples were obtained using Equation 1.

$$C = \frac{A}{P_{\gamma}(M \text{ or } V)T_c \varepsilon} \quad (1)$$

Knowing that:

M is the dried mass of the solid sample (Kilogram), V is the water sample volume (in Liter), P<sub>γ</sub> is the gamma emission probability (or branch ratio), T<sub>c</sub> is the counting time and ε is the detector efficiency.

### 3.4. Radiological parameters impact

To estimate and evaluate the radiological risk and the rate of danger to human health, some isotopic parameters were also calculated from the radioactivity measured on the ground in the study

area and in the laboratory. The absorbed dose rate  $D$  ( $\text{nGy h}^{-1}$ ) due to the activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  was evaluated using equation 2 according to international references.

$$D (\text{nGy } x\text{h}^{-1}) = C_U C_U + C_{Th} C_{Th} + C_K C_K \quad (2)$$

The parameters ( $C_U$ ,  $C_{Th}$  and  $C_K$ ) are the radioactivity concentration in  $\text{Bq kg}^{-1}$  and  $\text{Bq l}^{-1}$  for the solid and water samples from the study area, respectively, while ( $C_U = 0.462$ ), ( $C_{Th} = 0.604$ ) and ( $C_K = 0.0417$ ) are their dose conversion factors respectively [8]. The radium equivalent ( $R_{eq}$ ) activity index was determined using equation 3.

$$R_{eq} = C_U + 1.43 C_{Th} + 0.077 C_K \quad (3)$$

The external radiation hazards ( $H_{ext}$ ) and the internal radiation hazards ( $H_{int}$ ) were calculated using equations 4 and 5.

$$H_{ext} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \quad (4)$$

$$H_{int} = (C_{Ra}/185) + (C_{Th}/259) + (C_K/4810) \quad (5)$$

Radiation induced leukemia or gonadal damage is a possible occurrence when exposed to excess radiation, hence the need to estimate the annual gonadal equivalent dose (AGED). This was done using equation 6.

$$AGED (\mu\text{Sv y}^{-1}) = 3.09.C_U + 4.18.C_{Th} + 0.314.C_K \quad (6)$$

These parameters are the radioactivity concentrations in  $\text{Bq kg}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in the given samples.

The effective dose ( $H$ : health risk) due to the element radioactivity detected in the water samples was calculated using equation 7 [9].

$$H = R_a . A_i . C_f \quad (7)$$

$R_a$  is the radioactivity concentration of radionuclide in the water samples ( $\text{Bq l}^{-1}$ ),  $A_i$  is the annual intake ( $\text{y}^{-1}$ ) and  $C_f$  is the ingested dose conversion factor for radionuclides ( $\text{mSv y}^{-1}$ ), and varies with radionuclides and the age of individuals ingesting the radionuclides. Adding contributions from all radionuclides present in the study area, the total annual effective dose  $HE$  ( $\text{mSv y}^{-1}$ ) to an individual was found for the studied samples using equation 8.

$$HE = \sum (R_a . A_i . C_i) \quad (8)$$

According to the international protocol, during our study we adopted a subdivision according to different ages for good precision. The dose conversion factors used in the calculation and the annual effective dose were adopted from International Commission on Radiological Protection [10,11,12] for nine age groups (Table 2).

## 4. Results and Discussions

### 4.1. Spatial distribution of the radioactivity in the Dams

The concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  activities for liquid and solid samples are shown in Figures 3 and 4 respectively. The mean activity concentration measured for the water samples in the upper dam (Tunisia) was 1.74, 0.07, and 95.8  $\text{Bq l}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively. The mean concentration measured on the lower side of the dam was 1.70, 0.066, and 93.4  $\text{Bq l}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively. The mean activity concentrations follow the order  $^{40}\text{K} > ^{238}\text{U} > ^{232}\text{Th}$ . The mean activity concentration measured in the sediments collected at the upper side of the Sidi Salem dam (Tunisia) was 2.67, 0.18, and 197.87  $\text{Bq kg}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively. Concerning the Algerian Dam (Ain Dalia), the mean concentration measured on the lower side of the water dam was 1.9, 0.09, and 131.43  $\text{Bq l}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively. The mean activity concentration follows the order  $^{40}\text{K} > ^{238}\text{U} > ^{232}\text{Th}$ . The mean activity concentration measured in the sediments collected at the upper side of the dam was 4.34, 0.27, and 287.61  $\text{Bq kg}^{-1}$  for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively. It was

observed that the activity measured in the sediments was higher by a factor range of 1.57, 2.64, and 2.1 for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively, than what was measured in water samples in Sidi Salem (Tunisia). In the Algerian territory (Aïn Dalia Dam), it is the range of 2.3, 3, and 2.2 for  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  respectively, then what was measured in water samples. This is not surprising since sediments will transfer part of the radionuclide contents to the water and the Algerian constitute the origin of this natural radioactivity by diapirism. The estimated radiological impact parameters are presented in Table 1. The absorbed dose rate ( $\text{nGy h}^{-1}$ ) for sediment samples was below the word standard limit value (Table 1).

The estimated annual gonadal equivalent dose (AGED) is also less than the world limit of  $1 \text{ Sv y}^{-1}$ . The calculated absorbed dose for water was used to further calculate the total annual effective dose HE ( $\text{mSv y}^{-1}$ ) as shown in Table 2. The total annual effective dose obtained for all age groups was below the permissible reference limit [12]. This scientific report shows that this radioactivity contamination may be dangerous by the cumulative effect in the long term (depending on the socio-economic in this transboundary basin) and depending also in the future regional and global scenario in the North Africa basin and its relationship with the Mediterranean Basin.

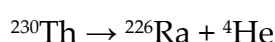
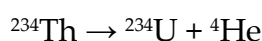
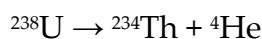
Aïn Dalia Dam (Recharge area-Algeria) and Sidi Salem Dam (Discharge area-Tunisia) reveals significant variations in radioactivity levels. The results indicate that the highest concentrations were observed in the lower part of the dams (sediment parts) which can be attributed to the organic matter and the clay minerals adsorption of the radioactivity [6]. The dead area (stagnation and compact zone in the bottom part) in the dam is composed by the stratified/compacted sediments. In this part depending to the drainage network input from the mount's erosion (clay, gypsum, sand, limestone/dolomite...). That's why the nearest dam (Aïn Dalia) from the sources of the natural diapiric radioactivity (salt, clay, sandstones...) from the Algerian Atlas is very enriched by this natural diapiric and metamorphic radioactivities. The decrease of the radioactivity rate from the Algerian territory to the Tunisian territory is due to the process of dispersion of the sediments (contact air/ water/rock interactions) in many effluents of the drainage network of Mallègue-Majerda basin (Tuniso-Algerian transboundary basin) [14, 15].

This radioactive distribution can be influenced by the tectonic activity, rock types, aquifer intercommunications, residence time, rain/wind rate, climatic parameters, and direction of the rain/wind, which can transport the radioactive particles and disperse them in different directions in the Majerda basin from the Algerian Atlas to the Mediterranean Sea (Tunis Gulf) (approximately within a radius of 300 km with a wind speed which oscillates between 20 and 100 km/h which depends on regional and global climatic conditions). The tectonic impact has a good role in the rechargeability/contamination of the aquifers and in the intercommunication between them in the study area [16, 17].

These dams (surface water and soil) show significant radioactivity variability. High levels of total radioactivity are generally in the range of  $0.21 \mu\text{Sv}$  to  $0.28 \mu\text{Sv}$  in the soil in Aïn Dalia dam and it increases with the water depth. From this recharge area (Algerian area) to Sidi Salem Dam (Tunisian discharge area), the levels of the total radioactivity are generally in the range of  $0.2 \mu\text{Sv}$  to  $0.23 \mu\text{Sv}$  in the soil and it increases with the depth also. It is the cumulative impact on the water and on the soil. These higher values are mainly recorded in specific areas such as the cities of Tebessa, Souk Ahras, Annaba, Jendouba, El Kef, and Beja, which are associated with the mining and ore industry and also with the presence of the diapiric and the metamorphic rocks in the high altitude (1,500 -2,800 m) [18, 19, 20, 21, 22].

These values are often observed in regions close to the surrounding mountainous areas, where the geological characteristics and the agriculture/industrial activities may play a role in the increase of radioactivity in the soil. These variations in radioactivity levels reflect the complexity of the geology, tectonic, environmental processes and socio-economic of North Africa area (Mallègue-Majerda basin of the Tuniso-Algerian transboundary basin). It is important to note that the radioactivity values mentioned are indicative and may vary depending on many factors, including local geological, geomorphological, climatic conditions, soil erosion and human activities. In this context, the study area is climatological under the influence of two types of air masses: (1)-

Mediterranean origin and the transport engine is the north/south direction wind, any radioactive activity (enrichment zone) in the southern European countries can be transported into this direction (to North Africa and the western part of the Middle East). (2)- The second air mass is desert/Atlantic, and the direction is South/North and West/East (to North Africa and the western part of the Middle East). All extraction activities from uranium mines as is the case in Niger and other countries in Central Africa will be transported by the wind and will be discharged in the study area (2000 Km) and in the rest of the MENA region countries (3000 to 3500 Km). The radioactivity disintegration can be resume in these equations:



Regional and global climate change has a key role in the balances and imbalances of ecological and environmental ecosystems. The conceptual model of the hydrodynamic of the surface water in these dams vs. the regional/global climate variation (arid/humid) (Figures 2 and 3), shows that the organic matter (OM) and the clay minerals have a good role in the radioactivity adsorption, in the immobilization of the radioactive isotopes and in the cosmic radiation. Combined with the influence of the irregularity of the flooding stage due to the regional and global climate variability in North Africa, the increasing arid paleo-climate played a very important role in decreasing the radioactivity by reducing the supply of clay minerals (adsorption/release), radioactive materials, and other minerals. This situation has also benefited primary paleo-productivity and anoxic conditions; their integrated effect could induce sapropelic "OM" accumulation (preservation and/or degradation). Therefore, the relationship between the OM content/clay minerals and the radioactivity is very complex; Rather than only assuming a positive correlation, its characteristics and formation mechanism need to be analyzed in a specific environment.

In the humid climate (Figure 2), the stormwater brings sediments rich in radioactive elements (clay, OM, sandstone, salts...), and rain showers recharge the dams and cause heterogeneity in the water column of the dams. This is why the level of radioactivity is high in the water column during wet periods. However, during the arid climate (Figure 3) when rain showers are almost null, the water column will be stratified (evaporation and density effects). From where the radioactive elements are deposited at the bottom part to their weight and the rate of radioactivity will be important in the basal parts of the dams. Moreover, during this period the shallow aquifer (alluvial and karstic) after the dams will be recharged by infiltration (period of deep-water contamination).



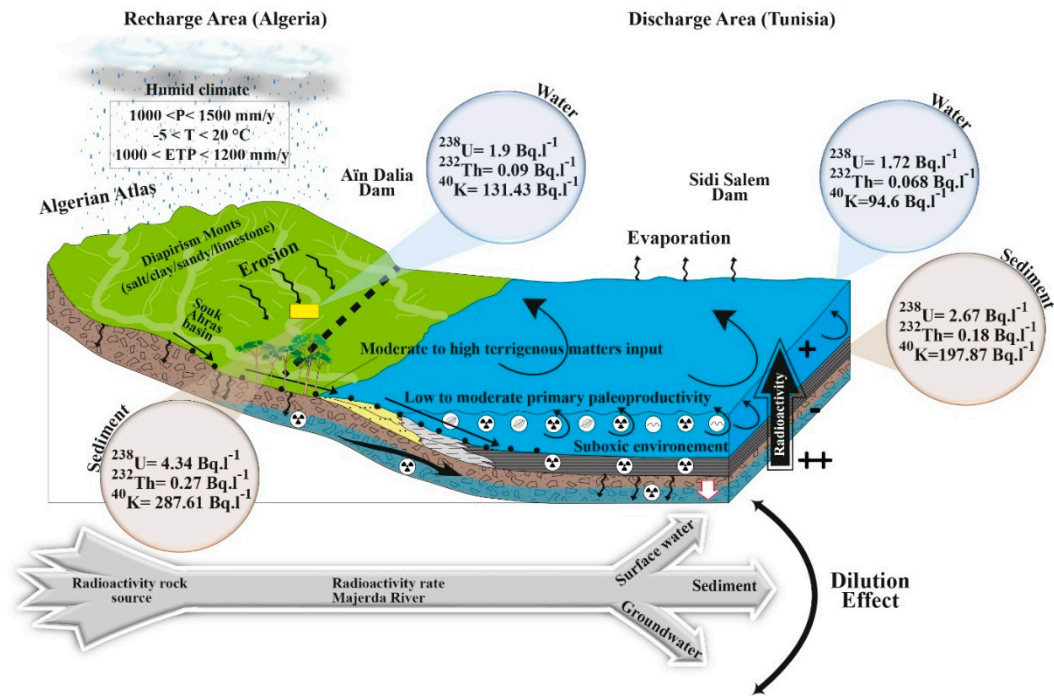


Figure 3. The hydrodynamic of surface water in the humid climate.

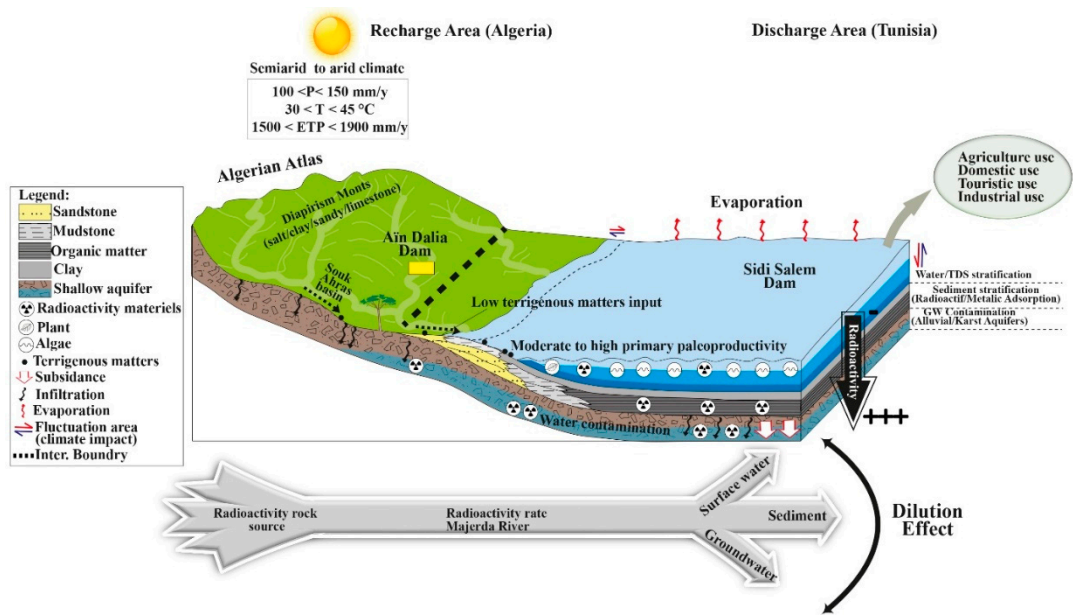


Figure 4. The hydrodynamic of surface water in the arid climate.

Table 1. Estimated radiological parameters for the sediments.

Location/country	D (nGy.h <sup>-1</sup> )	Raeq (Bq.kg <sup>-1</sup> )	Hext	Hint	AGED (μSvy <sup>-1</sup> )
Ain Dalia dam/Algeria	14.12	36.79	-	-	104.53
Sidi Salem dam/Tunisia	9.59	18.16	-	-	71.13
World standard value [8]	57.00	370.00	1.00	1.00	1.00 Svy-1

Table 2. Total effective dose (HE) for surface water samples (Sidi Salem Dam Tunisia).

Age Group	0-1	1-2	2-7	7-12	12-17	17-25	25-45	45-65	>65
Upper side (mSvy <sup>-1</sup> )	1.22	1.59	1.83	2.14	3.66	4.45	5.12	6.34	7.04
Lower side (mSvy <sup>-1</sup> )	0.84	1.09	1.26	1.47	2.52	3.06	3.76	4.12	5.02

Word Standard value [8]	0.12 mSvy <sup>-1</sup>
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In this present scientific report, it was noted that in all surface water samples in this tectonized transboundary basin in North Africa, the concentrations of <sup>238</sup>U were higher than those of <sup>232</sup>Th. The presence of uranium-bearing minerals associated with the upwelling of diapiric/metamorphic/sedimentary rocks together with limestone, dolomite, marl, salt, gypsum and clay commonly found in Tebessa, Souk Ahras and Annaba mounts (in Algerian Atlas) and in El Kef and Jendouba mounts (in Tunisian Atlas) can explain this observation in this report. The variability of the radio-activities can also be attributed to the oxydo-reduction conditions. Sometimes the fossil geothermal (40 Ky - 45 Ky) upwelling through the major faults increases the dissolution of these radioactive minerals.

<sup>40</sup>K is a radionuclide which abounds in the earth's crust and Ocean [13]. Its high rate in the surface water (dams and drainage networks) of the transboundary Majerda area may be due to the leaching of topsoil of nearby farmlands that employ inorganic potassium fertilizers to boot soil nutrients (orange, wheat, barley...). Using fertilizers in agricultural land, this isotope can percolate by infiltration and be leached in significant proportions in quantity into nearby vulnerable aquifers (alluvial and karstified aquifers). The rational safety of drinking water is an important water quality parameter of concern [8]. Most countries based their rational risk regulation and guidelines for drinking water on the UNSCEAR reports [8] and WHO limits [11]. The <sup>40</sup>K and <sup>238</sup>U rates obtained in this present study were significantly higher than UNSCEAR and WHO world average limits of 10.0 and 1.0 Bq.l<sup>-1</sup> respectively for drinking water. That of <sup>232</sup>Th is near the limit of 0.1 Bq.l<sup>-1</sup>. However, the values of the radioactivity in the soil show severe contamination and can be very dangerous for human health. From the radiological point of view, the accumulation of radionuclides <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K due to the ingestion of water from Majerda dams could present a low dose radiological risk of longer-term effects on health status of the Majerda population (the higher agriculture land in Tunisia and in North Africa, as it has been known for a long time until today: *Matmour Romal/Carthage/Tunisia*). When compared with other studies from some parts of the world (Table 3), it was found that the mean activity concentration of <sup>40</sup>K and <sup>238</sup>U was greater than those observed in water samples from Port Harcourt (Nigeria) [23]; Kuala Lumpur (Malaysia) [24], Ghana, Nigeria, Egypt, Makkah (Saudi Arabia) [25] and Gafsa (Tunisia) [6]. <sup>232</sup>Th concentration is lower than that found in some of these locations except for the value 0.12 Bq.l<sup>-1</sup> [25] observed in drinking water from KSA.

The <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K concentrations observed in the study area (transboundary basin Tuniso-Algerian basin) are also compared with those reported for other regions of MENA region (North Africa and Middle East) and worldwide average values in Table 3. However, the mean value of activity concentration of <sup>238</sup>U for the Tunisia zone was higher in the southern part of Tunisia (phosphate/petroleum mining basin) [6]. But lower in the Majerda basin (study area) when compared to the average values reported for the Middle East regions (KSA, Qatar, Bahrain, Emirate, Kuwait...) and the worldwide average values, the overall mean value for the entire study region was similar to these reported values.

Aim, the individual mean values of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K activity concentration for each zone as well as the overall mean value were lower when compared to the average value reported for the Middle East and the world. It is interesting to observe that the <sup>238</sup>U activity is higher in the soil when compared to the <sup>232</sup>Th concentration in the proposed uranium-mining region (cumulative impact).

**Table 3.** Comparison of specific activity of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K with other studies from different parts of the words.

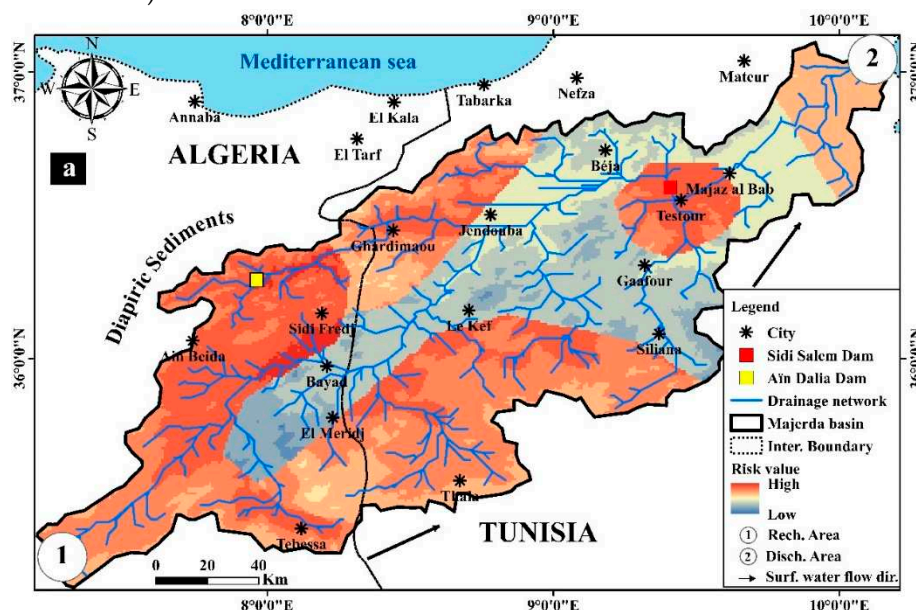
Region/Country	<sup>238</sup> U, Bq kg <sup>-1</sup>	<sup>232</sup> Th, Bq kg <sup>-1</sup>	<sup>226</sup> Ra, Bq kg <sup>-1</sup>	<sup>40</sup> K, Bq kg <sup>-1</sup>	Reference
Sidi Salem Dam (Water)/Tunisia	1.72	0.068	-	94.6	Present study
Sidi Salem Dam (Sediment) /Tunisia	2.67	0.18	-	197.87	Present study
Ain Dalia Dam (Water) /Algeria	1.9	0.09	-	131.43	Present study

Ain Dalia Dam (Sediment)	4.34	0.27	-	286.61	Present study
/Algeria					
Gafsa Phos.Rock/Tunisia	702	75	911	90	[6]
Tunisia	-	29	821	32	[26]
Egypt	686	5.7	656	68.6	[13]
Morocco	-	20	1600	10	[27]
Algeria	-	64	619	22	[28]
Saudi Arabia	-	17-39	64-513	242-2453	[24]
Germany	-	15	520	720	[27]
USA	-	49	780	200	[29]
India	-	65	120	2624	[28]
Jordan	-	2	1044	8	[26]
Kuala Lumpur-Malaysia	-	1.2	-	35.1	[24]
Buruta-Nigeria	-	26.9	-	61	[23]
Oman	-	2.26	20.49	44.83	[30]
Kuwait	-	6	36	227	[31]
Iran	-	17.61	14.96	361.6	[32]
Turkey	-	9.0	12.2	157.7	[33]

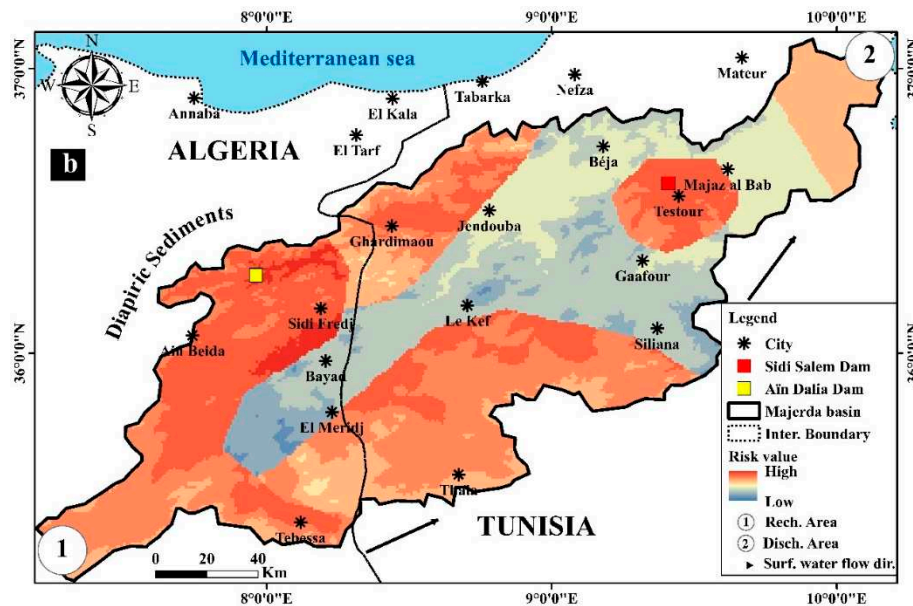
High U, Th, and K rates have been detected in surface water associated with diapirism/metamorphic/sedimentary rocks of the Algerian Atlas. These high concentrations are attributed to uranium minerals in granites and uranium minerals in pegmatites associated with these rock types of the Tuniso-Algerian Atlas and maybe also the result of the combination of these regional rocks' origin with the atmospheric circulation (Mediterranean origin from the north and Atlantic/desertic origin from the south).

#### 4.2. Risk map of the radioactivity distribution in the study area

Through the combination of the 12 parameters (Altitude, tectonic/fracturation, slope, lithology, aquifer levels/thickness, radioactivity rate, drainage network, surface/groundwater flow direction, rate/direction of rain and wind), we can establish the risk map (Figure 5a,b). The most significant values are observed at Ain Dalia Dam located near the diapirism outcrops (Algerian territory) and also at Sidi Salem Dam (Tunisian territory) by cumulative effect and finally in the final outlet (Tunis Gulf of the Mediterranean Sea/Carthage Sea) with average values. While the lowest values are observed in high altitudes in the study area. The drainage network with the permanent surface water has an important role in the transportation of the radioactive sediment and also has a role in the dilution of this radioactivity from the source of radioactivity (diapirism sediment) to the Tunis Gulf (cumulative sediments).

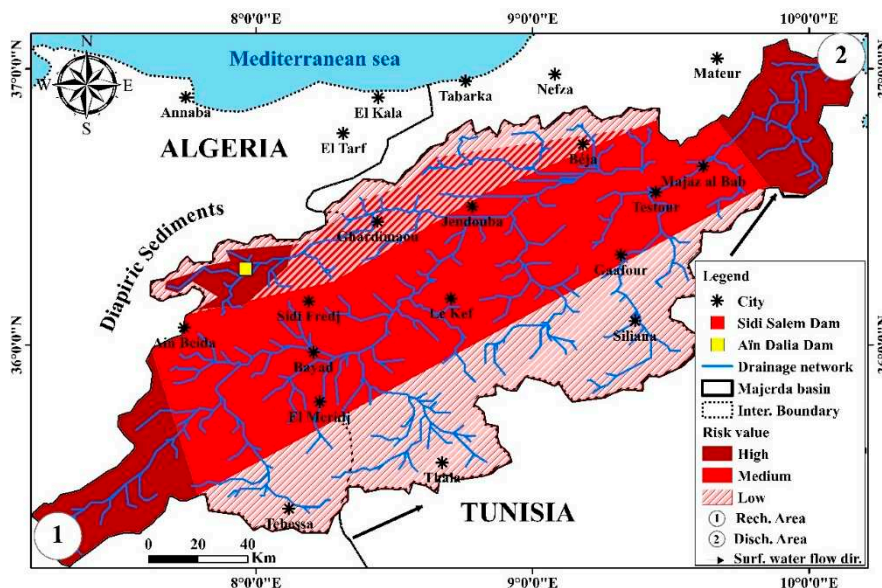






**Figure 5.** a-Risk map of the radioactivity spatial distribution in the study area with the drainage network; b- Risk map of the radioactivity spatial distribution in the study area without the drainage network.

The following map (Figure 6) shows that the highest levels of radioactivity are observed at the level of the diapirism rocks which constitute the source and the origin of the contamination in the Algerian territory (Transboundary Algero-Tunisian Atlas). We also see that the outlet of the watershed (Tunis Gulf) is marked by very high levels of radioactivity. This is essentially due to the cumulative effect. The lowest values are observed at high altitudes, generally carbonates, whose the radioactivity rate is low.



**Figure 6.** Health risk map of the radioactivity impact in the study area.

#### 4.3. Spatial distribution of the toxic heavy metals in the study area

The result of the concentrations of the toxic heavy metals (Fe, Pb, Zn, Ni, Cr, Cu and Cd) in the surface waters of the two dams of the Majerda River is also presented in Table 4. The concentrations of Fe, Pb, Zn, Ni, Cr, Cu and Cd in surface water range from 5430 to 9700 mg/L, 0.022 to 0.168, 0.018 to 0.142, 0.065 to 0.366, BDL to .0351, BDL-0.071 and BDL-0.048 mg/L respectively. The average concentration of heavy metals follows the trend Fe > Pb > Zn > Ni > Cr > Cu > Cd. This command

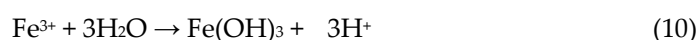
indicates the dominance of Fe, Pb and Zn in surface water samples from the Ain Dalia dam (Algeria). The average concentration level of Fe (7565 mg/L), this value is much higher than the standard values (0.3 mg/l) it is almost 25,217 times, Cu (0.0225 mg/L) was found to be below 2.00 mg/L of the WHO recommended limit. Ni (0.086 mg/L), Cr (0.061 mg/L) and Cd (0.009 mg/L) were slightly above the WHO recommended levels of 0.07, 0.05 and 0.003 mg/L respectively for drinking water [11], the Pb (0.218 mg/L) was found to be above the WHO authorized limits of 0.01 mg/L.

The implication of drinking such contaminated water may be in retarding certain biological and/or physiological processes. The relatively higher level of Pb and Ni in the water samples may be due to the anthropogenic pollution (mining extraction) along the Majerda River which feeds the two dams in this transboundary basin. Runoff from mines, dumps, and agricultural practices visibly present along the road and around the riverbank are plausible sources of heavy metal pollution from the river dam. There is also evidence of rock weathering and water-rock interaction from mineable and abandoned quarries as another source of metal pollution. Fertilizers and other agrochemical applications are a major contributing factor to the ubiquity of Pb in surface water bodies [9]. Lead is mainly used in the manufacture of lead batteries, it remains in landfills near rivers and streams and can serve as a source of lead pollution. This can diffuse through soil pores into groundwater and surface water [34 - 35].

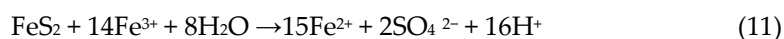
A high Pb rate in surface water from dams or through a cumulative effect can cause serious health complications such as poor memory, social disturbances, and reduced cognitive ability, while long-term exposure can lead to anemia [36]. Ni and Fe are found to be almost ubiquitous and very abundant elements in the environment (diapiric, meta-magmatic, and metamorphic rocks) [37]. The cropland is the main occupation of the inhabitants of the study area; therefore, a high concentration of metals in nearby streams and rivers is expected when the use of nitrogen fertilizers (synthetic nitrogen fertilizers and organic manure) is common [38]. According to Hamed et al. [6, 39], Cd is also present in a polluting form in phosphate fertilizers. The soil affected by sulfate minerals increased in acidity, in many cases reaching values below pH = 3.5 due to the acidity of the water (pH ≈ 5.2) and the pyrite sludge which, given its heterogeneity, ranged between 3.8 < pH < 5 [6]. The acidification of soils was aggravated by the oxidation of the sludge at the surface and even continued after the sludge removal tasks. The oxidation of the sulphides involves oxidation, hydrolysis, and hydration processes that Stumm and Morgan [40] summarized in Eq. (9).



This reaction starts with the release of  $\text{Fe}^{2+}$  and under oxidizing conditions is converted into  $\text{Fe}^{3+}$ . When soil pH is above 4, then  $\text{Fe}^{3+}$  precipitates as iron hydroxide and the pH becomes more acidic (Eq. (10)).



However, when soil pH remains under 4, the  $\text{Fe}^{3+}$  can oxidize the pyrite. This reaction is faster and can generate more acidity (Eq. (11)).



The concentration of these metals in solid samples is almost 20 times that in water, this is explained by the cumulative effect and by the presence of organic matter and fine clay sediments which are fixators of these toxic metals. In the polluted area by these metals can provoke many serious human illnesses.

**Table 4.** Specific Activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and the toxic heavy metals (Fe, Pb, Zn, Ni, Cu, Cr, Cd) in dams' surface water and sediments.

	$^{238}\text{U}$	$^{232}\text{Th}$	$^{40}\text{K}$	Fe	Pb	Zn	Ni	Cu	Cr	Cd
<b>Dalw</b> <sup>2</sup>	1.90 ± 0.24	0.09±0.01	131.43±1.03	5.430	0.0980	0.087	0.024	0.063	0.015	0.025
<b>Dals</b> <sup>3</sup>	4.34±0.05	0.27±0.05	287.61±3.34	201.9	5.88	5.046	0.216	0.441	0.06	0.20
<b>Dtnw</b> <sup>4</sup>	1.72±0.01	0.068±0.01	94.6±1.04	9.700	0.065	0.061	0.018	0.043	BDL <sup>1</sup>	0.130
<b>Dtns</b> <sup>5</sup>	2.67±0.01	0.18±0.012	197.87±2.01	136.7	3.41	3.22	0.182	0.213	BDL <sup>1</sup>	0.15



WHO limits	1.0	0.1	10.0	0.01	0.07	2.00	0.05	0.003
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<sup>1</sup> BDL: Below detection limit. <sup>2</sup> Dalw: Dam Algerian water. <sup>3</sup> Dals: Dam Algerian sediment. <sup>4</sup> Dtnw: Dam Tunisian water. <sup>5</sup> Dtns: Dam Tunisian sediment.

5. Conclusion and recommendations

The radioisotopes of <sup>238</sup>U, <sup>232</sup>Th and their decay descendants as well as the single non-series <sup>40</sup>K are transported to water by leaching and by infiltration in the vadose zone. This specific study will serve as a witness and index to carry out any environmental health monitoring in the transboundary basin of North Africa (Tunisian-Algerian zone) and why not in other similar area. The derived result will serve as a transposable study in other areas to verify any geochemical levels of surface and underground water resources (drinking water from dams, hill lakes and shallow aquifers). It will also provide future guidelines for providing safe drinking water sources to the community in the study area and may be transferable to similar locations in the MENA region (North Africa and the Middle East). The ingestion of water contaminated with radionuclides will result in the irradiation of human internal organs by alpha, beta, gamma and other types of radiation. This typical study showed that radioactivity in water give clear information from which the average radiation exposure from different sources can be estimated. Likewise, it showed that different types of fatal outbreaks (cancer) due to ingested radon from drinking water are equal to the total number of lung cancers due to radon inhalation. Continuous exposure to heavy metals and other chemical contaminants, even at low levels, could have a harmful effect on human health, especially at sensitive ages (under 17 years: the growing age of children).

Through this study, it is time to become aware of the control of transboundary water resources and to make master plans for quality and quantity control. To control the relationship between natural radiation and anthropogenic activities and the fact that human senses cannot detect low radiation (excluding the mine workers). International collaborations are essential to stop this type of quality degradation, especially of air and surface waters, which are in direct contact with the atmospheric column, ecosystem and human health. A global view on an international scale is crucial to fully understand the regional climatic situation in relation to the socio-economic development of the regions.

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**Conflicts of Interest:** The present paper is an original work, it is the first in the study area, and all the authors declare that they have no conflicts of interest. The authors jointly carried out this research study and they confirm that they are not associated with or involved in any profitable organization or company that has any financial interest. All authors have read and agreed to the published version of the manuscript.

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