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Radiological environmental monitoring of groundwater around phosphate deposits in North Africa-Southern Tunisia: Geochemical, Environmental and Health Factors

Monitoraggio radiologico e ambientale delle acque sotterranee in prossimità dei depositi di fosfati in Nord Africa-Tunisia meridionale: fattori geochimici ambientali e sanitari

Younes HAMED[a,b], Matteo GENTILUCCI[c], Hassen JALLOULI[a], Kaouther NCIBI[a], Riheb HADJI[d], Abderraouf JRABA[a], Ammar MLAYEH[e], Elimame ELALOUI[a]

[a] Department of Earth Sciences, Faculty of Sciences of Gafsa, University of Gafsa, Laboratory for the Application of Materials to the Environment, Water and Energy, Gafsa 2112, Tunisia - e-mail : *hamed_younes@yahoo.fr*; e-mail: *hassen.jallouli@fsgf.rnu.tn*; *nkawther@yahoo.fr*; *jrabaraoof@gmail.com*; *limam_aloui@yahoo.fr*

[b] Department of Earth and Atmospheric Sciences, Science and Research Building 1, University of Houston, 3507 Cullen Blvd, Room 312, Houston, TX 77204, USA

[c] School of Science and Technology, Geology division, University of Camerino, Camerino, Italy - e-mail: *matteo.gentilucci@unicam.it*

[d] Department of Earth Sciences, Institute of Architecture and Earth Sciences (IAST), Laboratory of Applied Research in Engineering Geology, Geotechnics, Water Sciences and Environment. University of Sétif 1, Setif 19137, Algeria - e-mail: *hadjirihab@yahoo.fr*

[e] Water Research and Technologies Center (CERTE), Tunisia - e-mail: *ammarmlayah17@gmail.com*

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Correspondence to:
Younes Hamed :
hamed_younes@yahoo.fr

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Riassunto

La contaminazione delle acque sotterranee è un problema globale che ha un impatto significativo sulla salute umana, sui sistemi ambientali e sul biota. L'atmosfera, i sedimenti e le acque idrotermali profonde della Tunisia meridionale contengono naturalmente isotopi radioattivi, provenienti principalmente dai depositi di fosfati dell'Eocene/Ypresiano, ed estratti dal grande serbatoio idrotermale del Nord Africa (sistema acquifero del Sahara nord-occidentale). Questi depositi di apatite possono essere arricchiti in Uranio-238 e nei suoi isotopi figli, che possono avere effetti nocivi sia sull'uomo che sull'ambiente. Il cambiamento climatico amplifica gli impatti devastanti delle radiazioni nucleari derivanti dall'estrazione mineraria e dall'industria, in particolare nella Tunisia meridionale. I noduli di fosfato contenenti elementi radioattivi provenienti dall'Eocene si sono ridistribuiti in formazioni geologiche più recenti, determinando una distribuzione capillare in tutta la regione meridionale della Tunisia. I risultati delle misurazioni relative alla radioattività naturale nel sud della Tunisia mostrano che i valori dei vari componenti (^{238}U, ^{232}Th, ^{226}Ra e ^{40}K) presentano variazioni significative a seconda della località (0,05 mSv/h < dose di radiazione gamma< 0,36 mSv/h nell'aria, 0,06 mSv/h < dose di radiazione gamma < 0,36 mSv/h nel suolo e tra 0,18 e 0,24 mSv/h nell'acqua sotterranea). Tuttavia, in prossimità dei depositi di fosfati, i livelli di radiazioni gamma e la radioattività complessiva sono elevati e superano gli standard fissati dall'Agenzia Internazionale per l'Energia Atomica, pari a 0,3 mSv/h. Ciò è dovuto alla presenza persistente di una nube di polveri contenente inquinanti metallurgici e radioattivi sopra le miniere di fosfato e le acque sotterranee idrotermali. Questi inquinanti, influenzati da parametri meteorologici come la velocità del vento, la temperatura, la nebbia e le precipitazioni, la circolazione delle acque sotterranee, vengono trasportati nelle regioni vicine. L'estrazione mineraria dai depositi dell'Eocene e le attività correlate hanno causato l'aumento dei livelli di radioattività nell'aria, nel suolo e nelle falde acquifere sul versante occidentale. L'intensificazione della contaminazione può manifestarsi in vari modi e può avere gravi impatti sulla salute umana e sull'ambiente.

Abstract

Groundwater contamination is a global problem that has a significant impact on human health and environmental/biota services. Southern Tunisia's atmospheric, sediments and deep hydrothermal naturally contains radioactive isotopes originating primarily from Eocene/Ypresian phosphate deposits and extracted from the great hydrothermal reservoir of North Africa (North Western Sahara Aquifer System). These apatite deposits can be enriched in Uranium-238 and its daughter isotopes. This can have harmful effects on both the human life and the environment. The changing climate amplifies the devastating impacts of nuclear radiation resulting from phosphate mining and industry sectors particularly in Southern Tunisia. The phosphate nodules carrying radioactive elements from the Eocene have been redistributed into more recent geological formations. This has led to their widespread distribution throughout the Tunisian southern region. The results of measurements relating to natural radioactivity in southern Tunisia demonstrate that the various components' values (^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) show significant variations depending on the location (0.05 mSv/h < gamma radiation dose< 0.36 mSv/h in the air, 0.06 mSv/h < gamma radiation dose < 0.36 mSv/h in the soil and between 0.18-0.24 mSv/y in groundwater). However, the gamma radiation levels and the overall radioactivity are high in the vicinity of phosphate deposits exceeding the standards set by the International Atomic Energy Agency of 0.3 mSv/h. This is due to the persistent presence of a dusty cloud containing metallurgical and radioactive pollutants above the phosphate mines and the hydrothermal groundwater. Influenced by meteorological parameters such as wind velocity, temperature, fog, and precipitation, groundwater hydrodynamic circulation, such pollutants are transported to neighboring regions. Mining of the Eocene deposits and related activities caused rising levels of air, soil and groundwater radioactivity on the western side. Intensified contamination can manifest in various ways and may have severe impacts on the human health and the environment.

Introduction

The worldwide radio-anthropogenic activities are once again causing damage to the environment. Activities like the French nuclear tests in Algerian Sahara (1959-1963), Chernobyl disaster of 1986, the Gulf war (1990-1991) and the Fukushima Daiichi nuclear disaster in 2011 caused a large quantity of radioactive waste to be generated and dumped into the ecosystem. Thus, the general population has been exposed to serious life-threatening disorders (cancer, cardiovascular illnesses, genetic modification, etc). Atmosphere, soil, water resources and alimentation food with toxic elements and particles propagation such as radionuclides (^{238}U, ^{232}Th, ^{224}Ra, ^{222}Rn, ^{222}Rn* and 40K) and aerosols (particles dust, gas and potential toxic elements (PTEs)…) caused many other health diseases (thyroid, breast and prostate cancers). The ecotoxicology contamination variability depends on sex, age, mining job, life in the mining basin and exposition time (8h to 24h) (Hamed et al., 2024b). In the environment, two primary sources of radiation can be distinguished. These are the natural ionizing radiations and the artificial ionizing radiations. Natural ionizing radiations originate from cosmic sources such as cosmic radiation from outer space that penetrates the atmosphere as well as the decay of naturally occurring radionuclides found in the ocean, soil, and rock (Al Zahrani et al., 2011; Chauhan et al., 2013). On the other hand, artificial ionizing radiations result from human activities. This includes nuclear power plants and the release of artificial radionuclides which contribute to environmental radiation levels. Additionally, industrial activities, such as mining, also add to this radiation source by emitting radioactive elements (Al Zahrani et al., 2011; Chauhan et al., 2013; Hamed et al., 2023).

There are numerous radioactive elements (radionuclides) found in the environment. Some of the most common radionuclide elements include Uranium-238, Thorium-232, Raduim-226, Radon-222 and Potassium-40. These elements undergo spontaneous disintegration into daughter nuclides with an associated emission of ionizing radiation in the form of alpha and beta particles and gamma rays (UNSCEAR, 2000; Duval et al., 2004). These daughter nuclides may either remain stable or continue to be radionuclides themselves undergoing further radioactive decay (IAEA, 2011; Islam et al., 2021).

The detection of the elements mentioned earlier is a critical phase. Various methods and techniques are employed to detect and quantify radioactivity levels in the environment. These include gamma-ray spectrometry, alpha and beta particle counting, and scintillation detectors. These detection methods enable to identify and measure some specific radionuclides present in soil, water, air, and biological samples (Wang et al., 2017). The radioactivity levels' accurate detection and measurement can help to identify and evaluate the potential radiation hazards. This information is essential for the understanding of the contamination context as it helps to assess the potential health risks for the exposed populations and, thus, guide appropriate risk management strategies (Zeng et al., 2021). Hazard detection also plays a crucial role in regulatory compliance by ensuring that radiation levels are within the acceptable limits defined by national and international radiation protection guidelines. In addition to hazard detection, it is important to consider hazard removal processes to mitigate the potential risks associated with radioactive or other hazardous potential PTEs and rare earth elements (REEs) (Colman and Holland, 2000; Khelifi et al., 2021a,b,2024; Hamed et al., 2023; Hidouri et al., 2024). Emerging materials have shown promise in the removal of radioactive heavy metals, such as uranium, from the environment. These materials, including various compositions, have demonstrated efficient adsorption and ion exchange capabilities (Zhang et al., 2022; Hazou et al., 2023). Their unique properties, such as large surface area, high selectivity, and chemical stability, make them potential

candidates for effective remediation of contaminated sites.

Producing 8 million tons of marketable phosphate. Tunisia plays a significant role in this context. Ranked as the fifth worldwide among producing countries, 85% of Tunisia's production is transformed into Phosphoric acid and fertilizers (Hamed et al., 2018). Considering the potential of nanomaterials for effective remediation and acknowledging Tunisia's prominent position as the fifth-largest phosphate producer globally, the country is exploring the extraction of Uranium from phosphate to fuel its future nuclear power plant. This strategic move aims to not only reduce the energy bill but also to foster socio-economic development at the international scale. Located 400 km southwest of the Tunisian capital city, Gafsa phosphate mining basin spans across 4,200 km^2 including the cities of M'Dilla, Metlaoui, Moulares, and Redeyef as noted by Hamami et al. (1880), Boujlel et al. (2008) and Hamed et al. (2008, 2010). Other studies have also identified new phosphate areas such as El Guettar, Jallabia, M'Ratta and Sidi Boubaker. Since the first discovery at Thelja Gorge in the late 19^{th} century, the Gafsa phosphate basin has become one of the world's most significant phosphate producers. According to Gafsa Phosphate Company-Tunisian Chemical Group (GPC-TCG), production increased from 200,000 metric tons/year in the initial excavation to over 8 million by 2009. However, Tunisian revolution of 2011 had socio-economic impacts on the Middle East and North Africa (MENA) regions resulting in oscillations in phosphate production (Hamed et al., 2008, 2023). The mining of Uranium-rich ore deposits such as phosphate rock, along with the growth of nuclear activities worldwide has amplified the possibility of human-induced radioactive pollution (WHO, 2011). Hence, safeguarding against such risks is crucial. Recognizing the importance and the danger of the matter, the Tunisian government has initiated a program aimed at examining the radiological environment in conjunction with the French Atomic Energy Commission and the International Atomic Energy Agency (IAEA). This study was initiated in the transboundary basin of Gafsa-Tozeur/Tébessa (Tuniso-Algerian basin) known, in particular, by its richness in phosphate deposits characterized by an average Uranium content of 35 ppm and by the biggest transboundary hydrothermal groundwater reservoir of NWSAS in North Africa. This study primarily aims to conduct in situ measurements and laboratory analyses of natural gamma radioactivity (specifically ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) across different geological levels within the Tuniso-Algerian transboundary phosphate zone and from deep hydrothermal groundwater of NWSAS. Additionally, it seeks to assess the naturally occurring radioactivity present in commercially available phosphate fertilizers within the Gafsa basin. The uranium distribution in the study area (rocks, soils and groundwater) provides information about the sedimentation and the oxido-reduction conditions, the determination of the activity ratios of $^{234}U/^{238}U$, the paleogeomorphology conditions, the groundwater interaction and hydrodynamic and the paleo-climatic conditions. The natural radionuclide uranium has three isotopes in the natural environment ^{234}U (0.0054%), ^{235}U (0.72%), and ^{238}U (99.27%) (Selvakumar et al., 2018). Particularly concentrated in the Gafsa region, Tunisian phosphate deposits are predominantly composed of carbonate fluorapatite (CFA) with notable concentrations of CaO and SiO_2 (Khelif et al., 2024; Garbaya et al., 2021; Hidouri et al., 2023, 2024). Extensively utilized as raw material for phosphoric acid production, these deposits also reveal the significant presence of various rare elements. This includes Cd, F, Li, Cr, Cu, As, Zn, Sr, Sb, V, and Eu which are identified at varying levels in residues from washing and processing operations within the phosphate industry. The detected elements' diversity these deposits complexity and their potential environmental impact. Importantly, these findings highlight the need for a comprehensive assessment of natural radionuclide content in Tunisia. This assessment is of paramount importance to evaluate the radiological implications and potential risks associated with phosphate extraction and processing in the Gafsa region. A thorough understanding of phosphate composition is crucial to guide resource management policies and minimize environmental consequences at both local and global scales. The study's focus extends to monitoring the radioactivity impact on air, soil, groundwater and human health contamination in the NWSAS transboundary basin.

Materials and Methods

Geographic location

The study area (Tunisia, North Africa, along the southern Mediterranean Sea) is situated in the Gafsa phosphate basin, encompassing the mining basin and its surrounding areas (Fig. 1). The study area has undergone climate shifts in climate ranging from arid to semi-arid conditions marked by seasonal fluctuations in climatic factors. The region is notably impacted by sandstorms originating from the Great Erg of the North African Sahara (SW/NE). This area experiences consistent exposure to two prevailing wind directions. These are the northeastern wind from marine sources (Mediterranean origin) and the southwestern wind from desert sources, commonly referred to as Sirocco (CRDA, 2021; Hamed et al., 2008, 2022a).

The study area is situated within a mining basin characterized by distinctive geomorphological features influenced by the collision of the Euro-African tectonic plates (Hamed et al., 2013, 2014; El Gayar and Hamed, 2017). These deposits' thickness varies based on several geological parameters. This includes climatic conditions, eustatic changes, and tectonic activities. Since the discovery of phosphate in 1880 by Hamami (Hamami et al.,1880) and in 1883 by Thomas (Thomas, 1893), phosphate extraction has primarily occurred in sub-surface quarries (10 - 40 meters deep) using traditional methods. Starting from the 1990s, phosphate mining has transitioned to open-pit operations with machinery. This resulted in the closure of numerous sub-surface quarries (Boujlel et al., 2008; Hamed et al., 2018; Besser and Hamed,

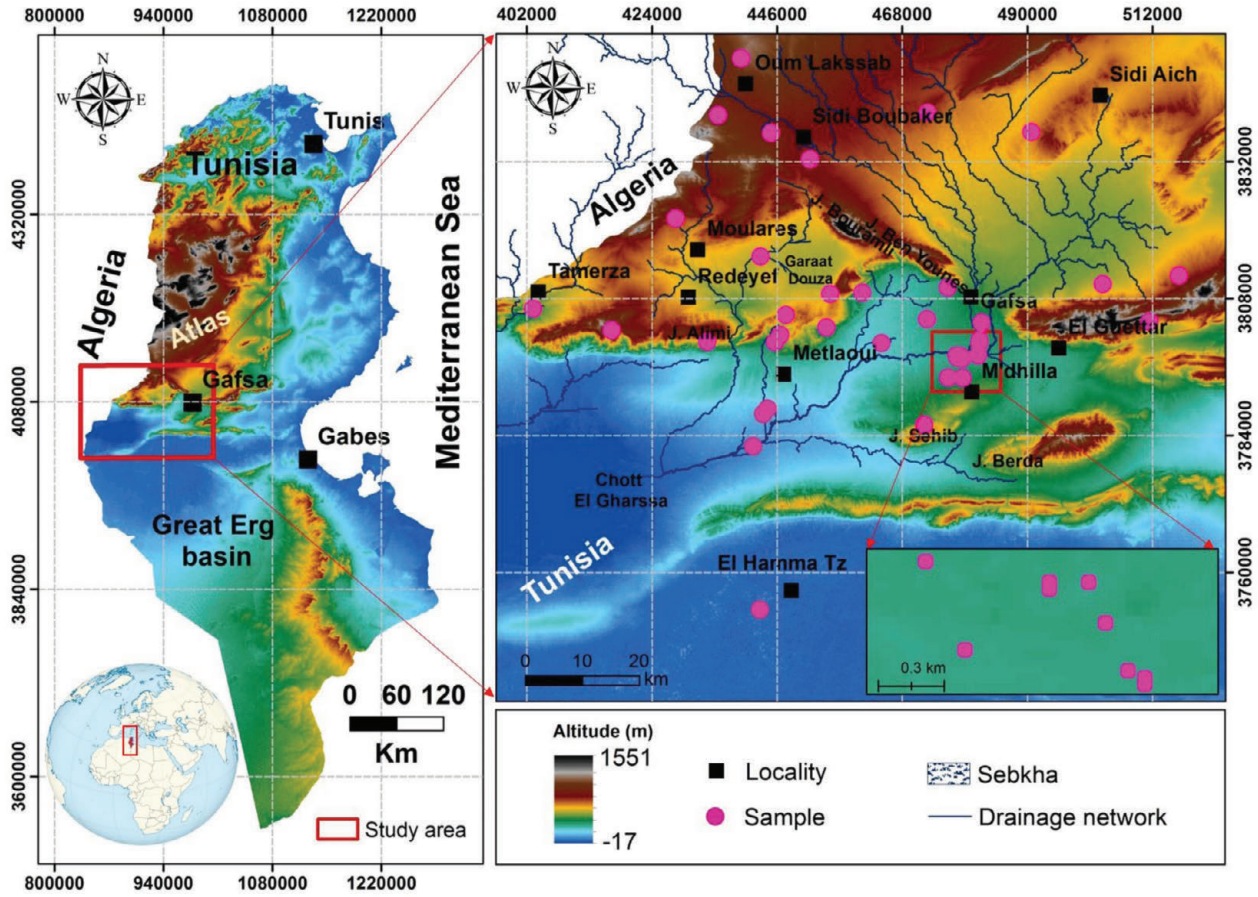

Fig. 1 - Location map of the study area (Phosphate Gafsa basin-SW Tunisia).

Fig. 1 - Carta dell'area di studio (Fosfati bacino di Gafsa - SW Tunisia).

2019; Melki et al., 2022).

Hydrogeology and tectonic setting

In southwestern Tunisia, the North Western Sahara Aquifer System (NWSAS) designates the superposition of multi-aquifer system. These include the Continental Intercalary (CI, sandy aquifer of Sidi Aich Fm.) and the Complex Terminal (CT, sandy, carbonate and dolomite levels of Segui, Beglia, Abiod/Berda and Zebbag Fms.) (Fig. 2). Given the desertic climate conditions, these reservoirs are slightly recharged. In fact, the recharge system represents about 1 billion m3/year. It essentially percolates in the Saharan Atlas piedmont plains in Algeria, the Dahar in Tunisia and Nefoussa Mounts in Libya (OSS, 2003) during the intense palaeorecharge (Humid phases and/or pluvial) from about 45 to 23.5 Ky (interglacial periods in the Mediterranean basin) (Guendouz et al., 1997; Zuppi et Saachi, 2004). However, the extension system and the layer thickness (200-1200 m) have facilitated the accumulation of considerable water reserves over the Pleistocene-Holocene humid climatic periods. According to OSS (2008), the paleo-groundwater "30-35 Ky" dates back to the Pleistocene and early Holocene under a cooler and humid climatic regime. This Pleistocene/Holocene period could be responsible for the piston-flow/mass effect periodic paleo-recharge of old groundwaters in the North Africa. In the last 70 Ka before present (MIS 2 and MIS 3, Würm period), three major humid phases have been identified: (i)- the Early Würm pluvial (MIS 3) lasting from 70 to 40 Ka before present, (ii)- the Middle Würm pluvial (MIS 3) lasting from about 32 to about 22 Ka before present and (iii)- the Late Würm pluvial (MIS 2), lasting from about 18 to 11 Ka before present (Zuppi and Sacchi, 2004 ; Hamed et al., 2008,2010,2014). These deeper reservoirs (1-3 Km) are hydrothermal water (35-92°C). Due to their residence time from the recharge to the discharge area, the processus of dissolution increases their mineralization (2-15 g/L) (Hamed et al., 2023). Piezometric studies show that groundwater converges towards the continental outlet of the transboundary depression of Chott area (Djerid/El Gharssa Chott). The Algero-Tunisian High Atlas and Nefoussa Mounts constitute the recharge area (Castany, 1981; OSS, 2003). About 8,800 water declared points, drillings, and springs exploit the NWSAS of this transboundary basin (3,500 from C.I and 5,300 from C.T). Per country water points are as follows: 6,500 in Algeria, 1,200 in Tunisia and 1,100 in Libya (OSS, 2003). Increasing drilling numbers and exploitation mode evolution indicate extremely strong withdrawal rates in the last thirty years: In 2000, the exploitation reached 2.5 billion/year (i.e. 1.50 in Algeria, 0.55 in Tunisia and 0.45 in Libya) compared to 0.6 billion in 1970 (OSS, 2003). At some

piezometers located in southwestern Tunisia (Tozeur region), an exceptional drawdown of the NWSAS water table was detected in 2008 reaching 8 m/year. This is due to the abusive and ab-normal exploitation in the Libyan territory following the construction of the Great Artificial River (GAR). If the trend observed in the countries continues, there will undoubtedly be serious reasons for concern for the Saharan regions future where we have already recorded the first signs of water resource deterioration. During the period 2000-2017, the number of drillings (legal and illegal) increased significantly in the 3 countries following political and socio-economic upheavals (OSS, 2003). The over-exploitation evolution of the NWSAS groundwater reservoirs in the agriculture and industrial (gas/oil and phosphate) sectors (0.6 to 20 billion m3/year) has profoundly modified the view that we can have from now on about this fossil hydrothermal groundwater abusive exploitation. These groundwater reservoirs are confronting a number of major and irreversible risks. These are the strong interference between the countries, the extinction of artesianism, the natural discharge depletion, the increased excessive drawdowns, water salinity increase, groundwater salinization inversion, perched aquifer neo-formed, springs extinction, subsidence land, groundwater hydrodynamic modifications, soil salinization, saline intrusion in some areas, groundwater budget incease and hydrothermal water radioactivity increase (OSS, 2003; Hamed et al., 2013; 2018, 2024a,b). Concerned by the system's future, the three transboundary countries, therefore, have to jointly search for some form of an optimal and concerted Erg basin management of NWSAS (international collaboration, control international system, international database collaboration etc). The major faults NW-SE and E-W directions (Gafsa-Tebessa fault, Negrine-Tozeur fault, Metaloui-Tabeddit, M'Dilla-Metlaoui fault, El Hamma-Agadir fault…) have a good role in the groundwater intercommunication. In the phosphate industrial wastewater discharge (continental depression of El Gharssa), the natural and the anthropogenic seismisity have an important role in the contamination of deep aquifers (intercommunication between the surface wastewater and the deep hydrothermal water/oil/gas reservoirs).

In situ Measurement

In 2023, two sampling campaigns were conducted in OIn this study, samples were analyzed using a radiologic TERRA detector which is known for its high sensitivity to climatic variations such as temperature (-20ºC <T<+50ºC) and humidity (relative humidity up to 95±3% at +35ºC), atmospheric pression (84 <A.P<106.7 KPa), as well as anthropogenic activities (IAEA, 2014). This state-of-the-art detector is capable of accurately detecting and quantifying

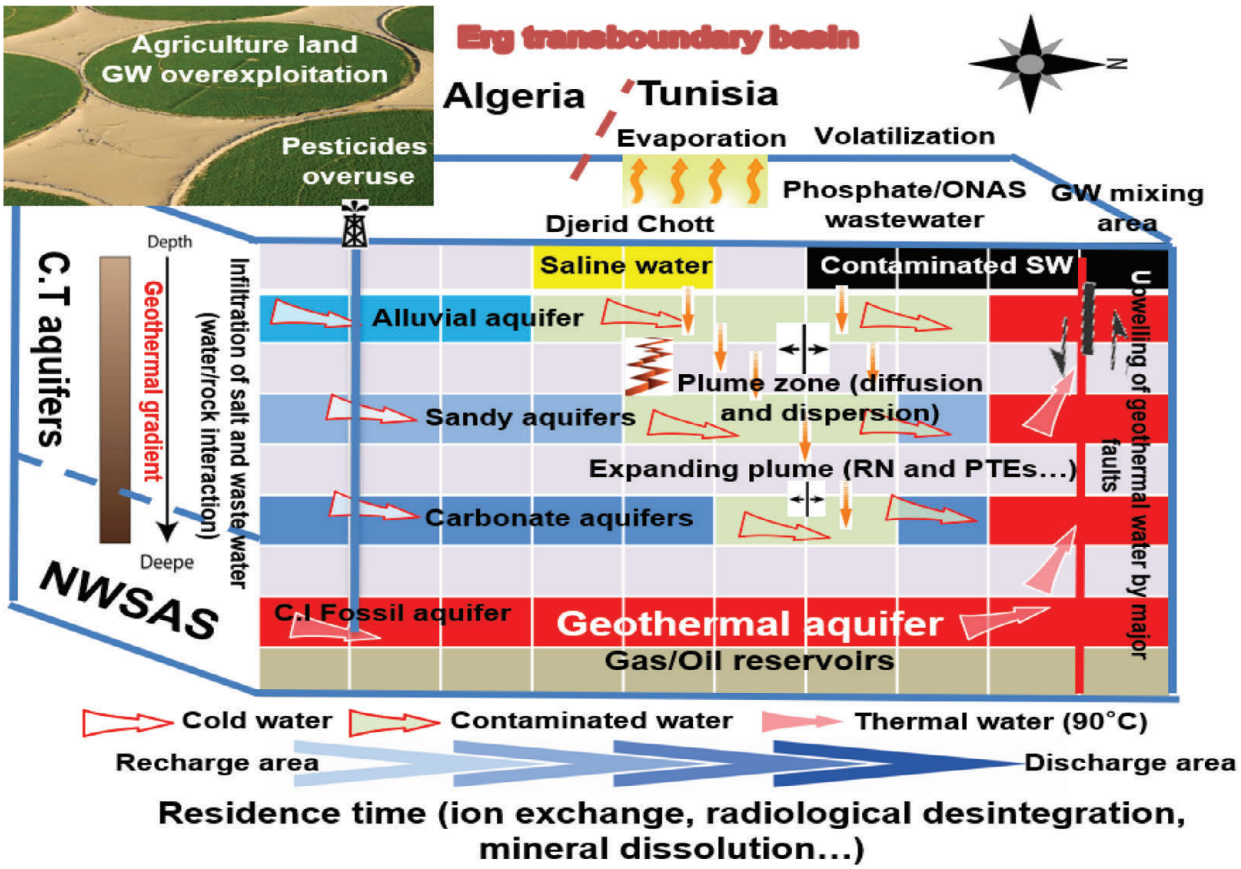

Fig. 2 - The different aquifers of the NWSAS transboundary basin.
Fig. 2 - I diversi acquiferi del bacino transfrontaliero NWSAS.

the cumulative levels of alpha and beta particles as well as gamma rays. This provides precise measurements of the diverse radioactivity forms present in the analyzed samples. It features high-resolution capabilities for air, sediment, and groundwater samples and includes a user-friendly display screen. Furthermore, it is shielded with a high-performance low-background lead shield at GeoLab-Aïn Azel-Sétif/ Algeria, ensuring the reliability of its measurements. Quality control (QC) procedures were applied using homemade control samples and reference samples provided by the IAEA (Environmental Monitoring Laboratory, Department of Energy, Ukraine). An IAEA reference multi-standard radionuclide source was utilized to calibrate the energy and efficiency of the detector (^{90}Sr + ^{90}Y and ^{137}Cs). In this study, a total of 76 samples were collected from the study area as follows: (i)- 43 samples were collected from various sources including crude phosphate, washed phosphate, solid sterile, phosphate releases, phosphogypsum, discharges from phosphate laundries, and uncontaminated soils that were not affected by phosphate pollution, (ii)- 33 samples were collected from deep wells from various aquifers of NWSAS and from different depths (depths ranging from 80 m to 2,900 m). To ensure that secular equilibrium was reached between ^{226}Ra and ^{222}Rn and their respective short-lived daughters, the samples were sealed and stored for a minimum of 28 days before measurements were taken.

Sampling

The pure phosphate activity concentrations of Uranium (^{238}U), Thorium (^{232}Th), Radium (^{226}Ra), and Potassium (^{40}K) were measured using a gamma-ray spectrometer with a high-purity germanium detector. Additionally, the radioactivity analysis of solid pure phosphate doped by lithium (HPGe) and cooled by the liquid-azote.

Activity concentrations (A)

The activity concentrations (A), in Bq/kg, of the gamma transitions emitted from the daughter radionuclides of the ^{238}U, ^{232}Th decay series (Fig. 3) and ^{40}K in the samples were calculated using the equation (Eq. 1) below with specific gamma lines corresponding to each isotope:

$$A = C_{net} / Iy(E)t\varepsilon_{abs}m \quad (1)$$

Where

A: samples activity concentration,

C_{net}: the net counts at a given energy transition,

Iy(E): the emission probability of gamma transition having certain gamma photons per disintegration for the respective radionuclide,

t: the counting time,

abs: photo peak efficiency at certain energy transitions,

m: mass of the measured sample (kg).

This equation takes into account the gamma transitions associated with each specific radionuclide to ensure accurate calculations.

Result and discussion

Phosphate rock radioactivity

Using gamma spectrometry, we conducted a thorough analysis of the phosphate rocks' radioactivity unveiling the preferential retention of radioactive elements such as ^{238}U and ^{232}Th in phosphoric acid while almost all of the ^{226}Ra were identified in the phosphogypsum by-product. The radioactivity rate is influenced by several factors including the phosphatic grain (pellets and their associated matrices), the maturity level of the humic substance "H.S" that is linked with the phosphate organic matter "P.O.M", fluoride, carbonate minerals, and the levels of clay present, all of which

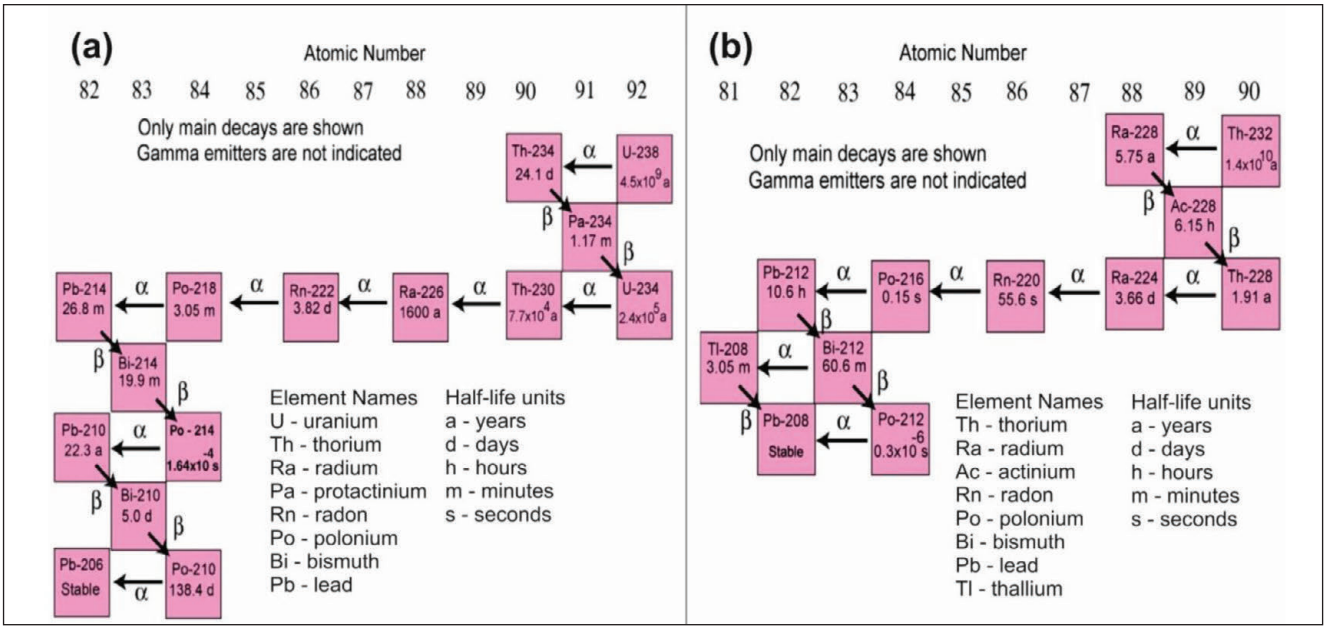

Fig. 3 - (a) The Uranium-238 decay and (b) the Thorium-232 decay chains (Duval et al., 2004).

Fig. 3 - (a) Catene di decadimento Uranio-238 e (b) Torio-232 (Duval et al., 2004).

can affect the rate of radioactivity.

Utilized as ores for the production of phosphoric acid, phosphate rocks are naturally radioactive as it is the case with many minerals. As a result, it is common to find them in the products and by-products of this manufacturing sector. Found that the radioactive elements ^{238}U and ^{232}Th remained preferentially in the phosphoric acid while almost all of the ^{226}Ra were found in the phosphogypsum. High-resolution gamma spectrometry measurements were performed at the GeoLab-Setif/Algeria yielding the following results (Tables 1, 2 and 3) in terms of average activity expressed in Bq/kg. It is not possible to directly identify ^{238}U in the spectrum because it cannot be detected by the (HPGe) detector due to its weak gamma energy. For this situation, we can determine the activity and the concentration of the ^{238}U in our samples using Phosphoric acid Spectrum X-rays before and after stability. The peak of ^{234}Pa m is used as a detection reference and from it we can determine all the other isotopes of Uranium (^{238}U), Thorium (^{232}Th), Radium (^{226}Ra), and Potassium (^{40}K) (Fig. 4 and Table 1).

Environmental impacts of phosphate transformation

The Tunisian Chemical Group (TCG) in M'Dilla/Gafsa plays a significant role in transforming Tunisian phosphate into phosphoric acid and agricultural fertilizers (TSP, DAP etc). This results in the emission of a diverse array of harmful substances. These pollutants encompass toxic gases (COx, SOx, NOx), particulate matter, potential toxic elements (PTEs), and radionuclides (such as U, Th, Ra, K, Sr, Cd, F, Pb, Zn, Li, Cu, etc.). All of which pose detrimental effects on both the environment and human health (Hamed et al., 2022b, 2024b). The carbon combustion during these processes releases CO and CO_2 gases. These are major contributors to greenhouse gas (GHG) emissions and global warming (Jatmiko et al., 2019). According to the USEPA (2021), approximately 65% of global GHG emissions are attributed to carbon dioxide, leading to diverse consequences of global warming and climate change. These consequences include the degradation of air, soil and water qualities as well as the occurrence of extreme weather events as highlighted by various researchers (Hamed et al., 2008, 2010, 2022b; Hartmann et al., 2000).

Generated during the petroleum and phosphate transformation activities, the NO_x (NO_2 and NO), and the SO_x gases (SO_2 and SO_3) are dispersed over long distances from the extreme south of Erg Basin (Libyan, Algerian, and Tunisian transboundary basins) to neighboring areas. When gases, especially NO_x, combine with atmospheric water or air humidity, they form sulfuric and

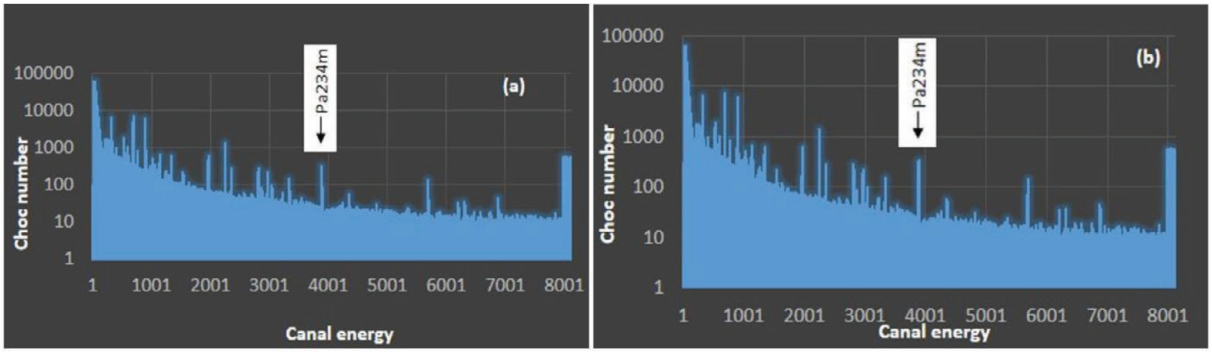

Fig. 4 - Phosphoric acid Spectrum X-rays of the fluorescence plumb armored, rays of naturals radionuclide's parents of ^{232}Th and ^{238}U (^{212} Pb, ^{214} Pb and ^{214}Bi) and the ^{40}K. (a)-Before stability (b)- After stability.

Fig. 4 -Acido fosforico, spettro a raggi X della fluorescenza a piombo, raggi dei genitori di radionuclidi naturali di ^{232}Th e ^{238}U (^{212}Pb, ^{214}Pb e ^{214}Bi) e del ^{40}K. (a)- Prima della stabilità (b)- Dopo la stabilità.

Tab. 1 - Comparison between activity concentrations average of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K of phosphate samples in the present study compared to published values worldwide.

Tab. 1 - Confronto tra le concentrazioni medie di attività di ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K nei campioni di fosfato del presente studio rispetto ai valori pubblicati a livello mondiale.

Region/country	^{238}U, Bq kg^{-1}	^{232}Th, Bq kg^{-1}	^{226}Ra, Bq kg^{-1}	^{40}K, Bq kg^{-1}	Reference
Gafsa Phos.Rock	702	75	911	90	This study
Tunisia	-	29	821	32	Olzewska-Wasiolek, 1995
Egypt	686	5.7	656	68.6	Islam et al., 2021
Morocco	-	20	1,600	10	Pfister et al., 1976
Algeria	-	64	619	22	Chauhan et al., 2013
Saudi Arabia	-	17-39	64-513	242-2,453	Al-Zahrani et al., 2011
Germany	-	15	520	720	Pfister et al., 1976
USA	-	49	780	200	Guimond et al., 1989
India	-	65	120	2,624	Chauhan et al., 2013
Jordan	-	2	1,044	8	Olzewska-Wasiolek, 1995

nitric acids which contribute to the formation of harmful acid rain. The effects of acid rain include soil erosion, disruptions in photosynthesis, stunted plant growth, and morphological changes in plant leaves (Mokadem et al., 2012; Mabrouk et al., 2020; Hamed et al., 2022a,b).

Additionally, ground-level ozone (O_3), along with nitric acid and organic compounds, contributes to the formation of photochemical smog in the study area (EPA, 2004). The latter is mainly triggered by the nitrogen oxides emitted from ore transformation factories. The stable climatic conditions with high summer temperatures (between 43°C and 52°C) in the regions of Gafsa/Tozeur/Kébili favor the concentration of these elements forming a peak of pollution. The atmospheric pollution and radioactivity degree depends on the weather situation in the study area (stable or instable weather). This atmospheric circulation is strongly controlled by the direction of the prevailing winds and the altitudes of the mountains surrounding the study area (Aurès transboundary border chain).

Moreover, the petroleum operations in the Tunisian regions of Tozeur and Kébili (within the Djerid petroleum basin) as well as in the southern part of the study area spanning Algerian and Libyan territories, along with the phosphate mining activities in the Gafsa basin of Tunisia and the Bir El Ater mine in Algeria, contribute to elevated particulate matter concentrations. These particles consist of fly ash, fumes, and dust derived from various sizes of apatite particles ranging from coarse to fine and even ultrafine particles. Petroleum and phosphate treatment activities (washing, transport, loading, and blasting) and phosphogypsum waste lead to the rejection of large quantities of different types of releases (GHG gases and other leaks). Potentially including PTEs and radioactive elements, exposure to particles has been linked to the incidence of respiratory and cardiovascular illnesses, cancer, infertility in young individuals, and genetic mutations leading to poor development in newborns (Hamed et al., 2010, 2014, 2024a,b, 2025; Khelifi et al., 2021a,b, 2024; Boschetti et al., 2025).

The increase in dust concentration can be attributed to the extraction of phosphate deposits by dynamite which involves open-air mining. The arid climate in the mining areas causes the persistent presence of a cloud of phosphate dust that varies in thickness depending on the topography and activities performed. For instance, M'Dilla, Métlaoui, and Moularés areas experience daily dust deposition of 18 g/m^2, 25 g/m^2, and 32 g/m^2 respectively under stable climatic conditions. The occurrence resulted in the pollution of nearby soil and potential contamination of the Mio-Plio-Quaternary (MPQ) unconfined shallow sandy and gravel aquifers which are highly susceptible to pollution in the region (Hamed et al., 2008, 2010, 2014; Ncibi et al., 2023). This in-depth analysis of emissions related to phosphate transformation highlights significant impacts on the environment and human health ranging from air pollution to the formation of acid rain. The diverse emissions, including toxic gases, rare toxic elements, and radionuclides, underscore the need for mitigation measures and strict regulations.

Soil and air radioactivities

The presence of nodular phosphate with radionuclides from the Eocene has been dispersed into younger formations and is widely distributed throughout southern Tunisia. Various samples, including raw phosphate rocks, water-washed phosphate rocks for commercial use, phosphate discharge after sieving, and TSPF recovery discharge, as well as non-phosphate rocks, soils such as marl, limestone, eolian sands, agricultural soil, and air, were collected from multiple regions in the phosphate mining basin. The findings from these samples are presented in Figure 5 and Table 2.

The gamma radioactivity levels distribution map in the mining basin (Fig. 5) shows a heterogeneous spatial distribution. It is strongly related to phosphate levels in the mountains. There is a good correlation with the thicknesses of the phosphate layers and especially the organic matter content (phosphate pellets) (Boujlel et al., 2008). According to this study, the levels of effective dose rate resulting from gamma radiation increase from the west of El Guettar region (0.31 mSV/h), the M'Dilla region side (0.33 mSV/h) (the open region on the Mediterranean Sea side) to the east on the high altitude side of the Aurès chain of the Tuniso-Algerian transboundary Atlas (location of the phosphate deposits around the Kasserine paleo-island), Métlaoui (0.33 mSV/h), (0.36 mSV/h in the phosphate cave of Moularés basin) and in Rédayef basin (0.34 mSV/h). These activity concentrations of radioactivity are higher than those of the world average (0.3 mSV/h). In the northern part of the study area (Sidi Boubaker region and north Gafsa basin), the values of radioactivity are low in the order of 0.06 mSV/h. This leads us to suggest a total absence of deposits of phosphate in these regions located in the north of this Aurès/Atlas Mountain (topographic or

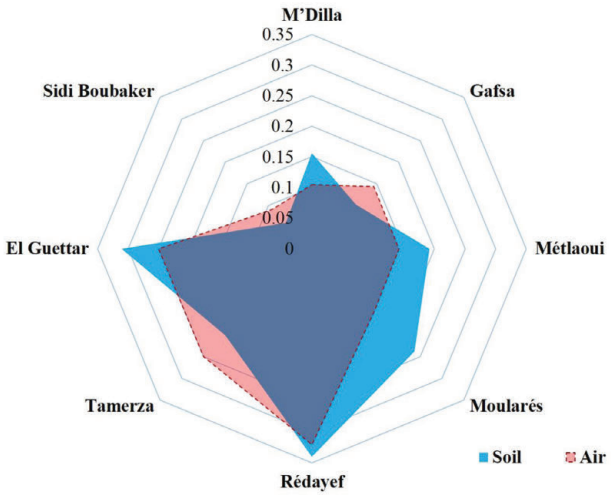

Fig. 5 - Radioactive measurements in the soil and air of the study area.

Fig. 5 - Misurazioni della radioattività nel suolo e nell'aria per l'area di studio.

Tab. 2 - Soil and air gamma radiation analysis in the study area.

Tab. 2 - Analisi della radiazione gamma del suolo e dell'aria nell'area di studio.

Region	Samples	X	Y	Soil Gamma Radiation (mSV/h)	Air Gamma Radiation (mSV/h)
M'Dilla	S1	478533	3797538.7	0.19	0.12
	S2	478532.93	3797509.1	0.12	0.11
	S3	478456.41	3797570.3	0.13	0.12
	S4	478354.68	3797786.8	0.13	0.11
	S5	478278.45	3797971.7	0.13	0.1
	S6	478099.49	3797940.7	0.12	0.08
	S7	478099.56	3797972.2	0.13	0.09
	S8	477537.58	3798065.9	0.18	0.09
	S9	477715.51	3797664.4	0.09	0.07
	S10	481345.33	3798118.5	0.09	0.05
	S11	481579.63	3800304.3	0.08	0.08
Gafsa	S12	476034.95	3809989.9	0.11	0.08
	S13	481969.7	3803846.3	0.11	0.1
	S14	478601.85	3794119.6	0.09	0.23
M'Dilla Lav	S15	476045.55	3794218.2	0.31	0.09
Gafsa	S16	481605.85	3800644.3	0.09	0.19
	S17	472394.31	3804424.2	0.09	0.19
	S18	464359.5	3800232.2	0.12	0.07
Métlaoui	S19	436344.23	3783346.4	0.15	0.15
	S20	434136	3778709.8	0.17	0.14
	S21	434034.66	3778865.7	0.21	0.08
	S22	445683.51	3800383.1	0.19	0.07
	S23	446559.58	3801671.8	0.33	0.32
	S24	447575.59	3805177.5	0.1	0.09
Moularés	S25	443118.07	3815400.2	0.1	0.36
	S26	454664.2	3803015	0.14	0.23
	S27	428184.68	3822043	0.36	0.22
Rédayef	S28	416930.24	3802477.6	0.34	0.32
Tamerza	S29	403122.69	3806245.5	0.2	0.25
M'Dilla Car.	S30	471870.56	3785939.3	0.33	0.25
El Guettar	S31	511513.27	3803906	0.31	0.25
Sidi Boubaker	S32	451951.06	3832463	0.06	0.25
	S33	444966.05	3837066.8	0.06	0.1
	S34	435669.79	3840147	0.06	0.07
	S35	439674.37	3850126.6	0.06	0.08
	S36	472482.77	3840601.6	0.06	0.08
	S37	490686.14	3837214.9	0.06	0.05
	S38	503280.33	3810544.9	0.06	0.05
	S39	516641.82	3812105.9	0.06	0.05
Moularés	S40	460980	3809061	0.33	0.05
	S41	455350.97	3808838.3	0.33	0.05
	S42	442981.67	3753566.7	0.1	0.05
	S43	433747.7	3800424.5	0.3	0.05

geomorphologic barrier) (Figs. 4 and 5).

The presence of natural radioisotopes, specifically Uranium and its progeny, is frequently linked to sedimentary deposits that are rich in organic matter and non-marine formations with an elevated concentration of Uranium and other PTEs. Quite high concentrations (^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) have been found in anoxic conditions in the presence of organic carbon. According to this study, that shows (^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) concentrations in sediments of the Eocene phosphate of the Gafsa basin, increased proportionately with organic concentration from 4 ppm at an organic content of 9% to about 32 ppm at an organic carbon concentration of 17%. Many of the highest reported concentrations of Uranium are found in phosphatic sediments of El Guettar, M'Dilla, Métlaoui, Moularés, and Rédayef phosphate mines (Fig. 6). Geochemically, (^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) tend to adsorb onto accumulating organic matter during sedimentation in anoxic areas. This is due to the geomorphological and the tectonic of the basin and to the diapirism and the magmatic/

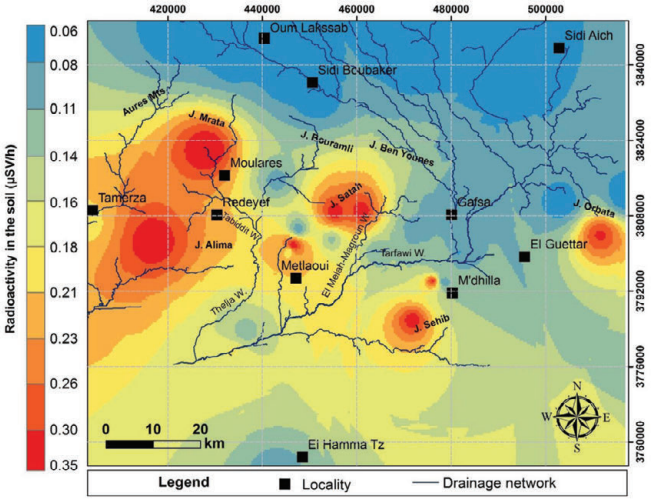

Fig. 6 - Spatial distribution map of soil radioactivity in the study area.

Fig. 6 - Carta della distribuzione spaziale della radioattività del suolo nell'area di studio.

metamorphic endogen activities in the study area.

The open-air extraction, as well as the contact with water during the washing process, combined with the unique geomorphological characteristics of the Métlaoui basin, contribute to significant values of radioactivity particularly in the air. These values are surpassing the established thresholds which typically range around 0.33 µSv/h. The spatial distribution of total gamma radioactivity (^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K) aligns with the distribution observed in the soil, albeit with lower values. This decrease in values, particularly for ^{226}Ra, can be attributed to various factors. This includes meteorological conditions such as air movement, wind patterns, temperature, solar radiation, humidity, and other environmental factors. Further studies are needed to provide concrete evidence of the dilution effect of 226Ra by meteorological factors (Fig. 7).

Contact with air and washing water causes disintegration

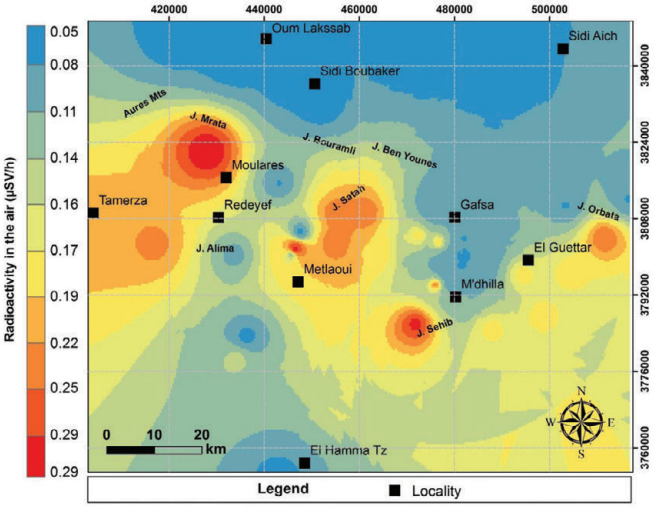

Fig. 7 - Spatial repartition map of the radioactivity in the air in the study area.

Fig. 7 - Carta di ripartizione spaziale della radioattività nell'aria nell'area di studio.

and volatilization of the harmful Radon-gas (^{222}Rn) isotope into the atmosphere. Several geographical sites in the study area were visited to monitor and to better understand the evolution of contamination following the flow of surface water from upstream to downstream in the catchment area (Figs. 8 and 9). Going towards the outlet of the hydrographic network of all these discharges (from phosphate washing and TSPF recovery), the levels increase again given that the soils of these rivers with the permanent flow by these wastewaters and the atmosphere are very enriched by these radioactive isotopes. Conversely, erosion and atmospheric circulation may lead to the dispersion of radionuclides particularly in this mining environment (phosphate and petroleum basins). Raw rock from the Eocene deposits has shown increasing levels of soil and air radioactivity ranging from the western maritime side of the Mediterranean Sea to the continental eastern side

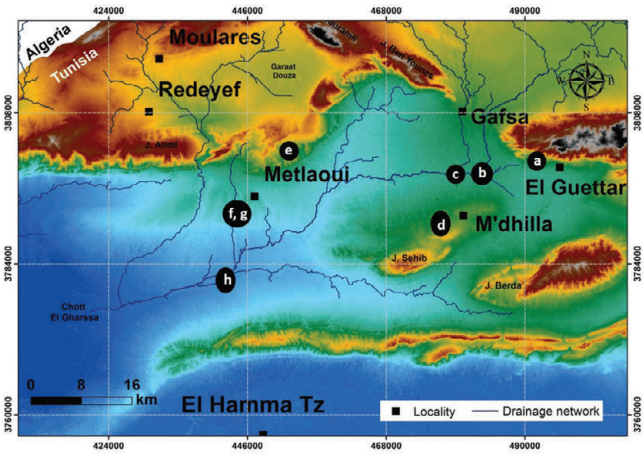

Fig. 8 - Location of the sampling for radioactivity measures in the study area.

Fig. 8 - Ubicazione del campionamento per le misure di radioattività nell'area di studio.

Fig. 9 - Photos showing the impact of the atmospheric and wastewater contamination in the study area (a)-direction of the atmospheric pollution from GCT to El Guettar basin blocked by Orbatta Monts, (b)-phosphogypsum stock impact, (c)- wastewater from phosphate valorization in El Melah river, (d)- wastewater from phosphate lavatory of M'Dilla basin, (e)- Phosphate rocks in the Oum Lakhcheb area (Métlaoui basin), (f and g)- Phosphate wastewater of Métlaoui lavatory (Thelja River) and (h)- The result of wastewater of phosphate and phosphogypsum mines (final outlet of all the discharge of phosphate from M'Dilla, Métlaoui, Moularés, and Rédayef regions).

Fig. 9 - Foto che mostrano l'impatto della contaminazione atmosferica e delle acque reflue nell'area di studio (a)-direzione dell'inquinamento atmosferico dalla GCT al bacino di El Guettar bloccata da Orbatta Monts, (b)-impatto dello stock di fosfogesso, (c)-acque reflue dalla valorizzazione dei fosfati nel fiume El Melah, (d)-acque reflue dal lavatoio dei fosfati del bacino di M'Dilla, (e)- Rocce di fosfato nell'area di Oum Lakhcheb (bacino di Métlaoui), (f e g)- Acque reflue di fosfato del lavatoio di Métlaoui (fiume Thelja) e (h)- Il risultato delle acque reflue delle miniere di fosfato e fosfogesso (sbocco finale di tutti gli scarichi di fosfato delle regioni di M'Dilla, Métlaoui, Moularés e Rédayef).

(around 120 km).

During sandstorms and under the effect of the south-north wind direction and average wind speed (50 km/h), aerosols and dust from the Tuniso-algerian transboundary phosphate basin (Gafsa/Tebessa) (unconventional uranium mine) can reach a great part of North Africa in 8 hours by the radio-toxic elements. The air/dust-mass-dynamics with the dominant south wind to north direction causes the dispersion of the toxic radionuclides over long distance and the contamination of vast territories of North Africa and Southern of Europe (Portugal, Spain, France and Italy).

Unconventional uranium dust + Atmospheric gases (CH_4 and N_2O (77%) + CO_2 (20%) + Argon (1%) + water vapor (0-4%) + trace gases: Ne, He, Kr, O_3 (0.01%)) + Rainfall from Southern Atlantic Ocean Circulation → UO_2N + UCO_2 + UO_2H + H_2O (acid rain water) + fine particles.

Another global atmospheric circulation of the wind from the south to the north is the main factor in the spread of radioactive dust from the conventional uranium mines in Niger (Central of Africa). The winds from the SW/NE directions can transport this dust to MENA region (North Africa and Middle East) and the Europe countries (Hamed et al., 2024a,b).

NWSAS Groundwater radioactivities

Uranium and thorium in sandy/limestones/dolomites rocks that constitute the North Africa great aquifer of NWSAS are highly soluble under the increasing of temperature (impact of the geothermal gradient: 30°C/Km - 10m<depth<3,000 m) conditions. Therefore, the controlling hydrodynamics for radionuclide release are dissolution at the water/rock interaction and alpha-recoil (ejection from the rock due to alpha-decay of the parent radionuclide). Uranium solubility is a strong function of the oxidizing or reducing nature of the geofluid. Uranium contents in NWSAS groundwaters are frequently more than the recommended limit for drinking water (20 µg/L) (Gascoyne, 1989; Hamlat et al., 2001). Uranium does not emit gamma radiation. Therefore, it is only hazardous if directly ingested or inhaled. Unfortunately, this is the case in the south of Tunisia. In fact, the geothermal waters of NWSAS are widely used in traditional and modern therapeutic baths (El Hamma of Gabes and Tozeur regions and Sidi Ahmed Zarroug of Gafsa region). Also, these waters are used in the domestic and agricultural sectors but after cooling as shown in the photos in Figure 10. The rate of the gamma radioactivity of the hydrothermal water from NWSAS in the El Hamma area (Tozeur region) is about 0.25 mSv/y (just outside of well), 0.18 mSv/y at the surface and 0.12 mSv/y at the air near the well. At the Sidi Ahmed Zarroug area (Gafsa region), the rate of the gamma radioactivity of the hydrothermal water from NWSAS is about 0.22 mSv/y (in the therapeutic station), ^{238}U= 1.05 Bq/L, ^{234}U=0.52 Bq/L, ^{228}Th=0.21 Bq/l, 228Ra=1.02 Bq/L and 226Ra=7.56 Bq/L (Fig. 10 and Table 3). Concerning the ^{40}K values, they are about (380 Bq/L). This high value is in relation with the metamorphic and evaporate rocks (KCl) dissolution during the upwelling of the hydrothermal water of NWSAS, atmospheric contributions rich in wind sediments from chotts and/or sebka and due to the excessive use of fertilizers in the agriculture lands.

Several international references (DOE, 2010; CEU, 2013; IAEA, 2013; WHO, 2022) are used to compare the doses found with the standard doses. Table 4 summarizs the dose limits for water established by different organizations and regulators which declare the total effective dose must not exceed 0.25 mSv/y for the drinking water (NRC, 2006, 2022).

We use these references which require different limits of annual effective dose equivalent limit established by different organizations. US.DOE requires the lowest value (0.04 mSv/y) and IAEA with a high value of 1 mSv/y. We can conclude that

Fig. 10 - Field radioactivity data of the geothermal groundwater of NWSAS in Southwestern Tunisia. A- Air gamma radioactivity, B- geothermal water circulation gamma radioactivity and C- surface geothermal water gamma radioactivity (December, 2022).

Fig. 10 - Dati sulla radioattività di campo delle acque sotterranee geotermiche del NWSAS nel sud-ovest della Tunisia. A- radioattività gamma dell'aria, B- radioattività gamma della circolazione dell'acqua geotermica e C- radioattività gamma dell'acqua geotermica di superficie (dicembre 2022).

Tab. 3 - Concentrations ^{238}U, ^{228}Th, ^{226}Ra (Bq/L) and gamma radioactivity (mSv/y) in shallow and deep groundwater samples in NWSAS transboundary basin.

Tab. 3 - Concentrazioni di ^{238}U, ^{228}Th, ^{226}Ra (Bq/L) e radioattività gamma (mSv/y) in campioni di acque sotterranee superficiali e profonde nel bacino transfrontaliero NWSAS.

Region	Sample ID	Gamma radioactivity mSv/y	^{238}U Bq/L	^{234}U Bq/L	^{228}Th Bq/L	^{228}Ra Bq/L	^{226}Ra Bq/L
M'Dilla	*GW1	0.18	0.19	0.83	0.24	2.05	7.95
M'Dilla	*GW2	0.16	0.2	0.75	0.22	2.00	7.98
M'Dilla	*GW3	0.22	0.22	1.22	0.1	0.84	4.48
El Guettar	*GW4	0.24	0.22	1.23	0.11	0.88	4.56
El Guettar	*GW5	0.19	0.20	1.09	0.1	0.78	4.49
El Guettar	*GW6	0.16	0.21	0.76	0.24	2.04	8.02
Gafsa North	*GW7	0.09	0.12	0.67	0.12	1.24	7.87
Gafsa North	*GW8	0.06	0.09	0.5	0.09	1.17	7.32
Gafsa North	*GW9	0.07	0.08	0.48	0.085	1.15	7.28
Gafsa North	*GW10	0.08	0.11	0.52	0.11	1.18	7.64
Metaloui	*GW11	0.14	0.2	0.61	0.11	1.21	6.98
Metlaoui	*GW12	0.15	0.19	0.58	0.12	1.24	7.09
Metlaoui	*GW13	0.18	0.23	0.66	0.14	0.96	1.87
Moulares	**GW14	0.08	0.1	0.32	0.13	0.52	0.89
Moulares	**GW15	0.07	0.11	0.33	0.16	0.66	1.05
Moulares	**GW16	0.09	0.12	0.42	0.18	0.79	2.07
Sidi Ahmed Zaroug	**GW17	0.24	1.05	0.52	0.21	1.02	7.56
Tozeur	**GW18	0.23	0.82	0.47	0.19	0.81	5.21
Tozeur	**GW19	0.26	0.98	0.41	0.2	0.72	4.45
Tozeur	**GW20	0.24	0.87	0.52	0.09	0.55	7.02
Sidi Boubaker	**GW21	0.10	0.09	0.31	0.04	0.42	2.63
Sidi Boubaker	**GW22	0.09	0.1	0.34	0.05	0.33	2.91
Oum Laksab	*GW23	0.07	0.08	0.31	0.02	0.29	2.78
Oum Laksab	*GW24	0.07	0.05	0.38	0.02	0.29	2.78
Oum Laksab	*GW25	0.08	0.04	0.35	0.01	0.31	3.21
Gabes	*GW26	0.26	0.29	1.04	0.13	0.2	0.98
Gabes	**GW27	0.29	0.32	0.98	0.12	0.14	1.02
Gabes	**GW28	0.28	0.24	0.78	0.1	0.16	1.12
Gabes	**GW29	0.31	0.33	1.05	0.11	0.12	ND*
Jerba	*GW30	0.06	0.04	0.28	0.02	0.24	3.01
Jerba	**GW31	0.24	0.23	0.66	0.11	0.14	1.14

Tab. 4 - Annual effective dose equivalent limit established by different organisations.

Tab. 4 - Limite annuale di dose efficace equivalente stabilito da diverse organizzazioni.

Organisation	Scenario	Limit on annual effective dose equivalent	Reference
WHO	Drinking water	0.1 mSv	WHO, 2022
European Commission	Drinking water	0.1 mSv	CEU, 2013
IAEA	Drinking water	1 mSv	IAEA. 2014
US DOE	Drinking water supplies operated by or for the DOE, not including naturally occurring radiation sources	0.04 mSv	DOE, 2010
US NRC	Criterion for license termination of nuclear facilities with unrestricted release, including the residual radioactivity from groundwater sources of drinking water	0.25 mSv	NRC, 2006, 2022

the annual cumulative effect in the phosphate mining basin exceeds these international references. The monitoring in real-time information (quantity and quality) promotes safety with the field of human health staff and all the population and the ecosystem of the mining basin.

Migration's paths of the radioactive contaminants

The impact of climate change on radioactivity in the air, rainfall, soil, vadose zones, and groundwater is also explored in this study. The authors hypothesize that the impact of climate change can be observed as disruptions to the natural recharge of the multi-aquifer system in the study region. This effect is usually more pronounced when the recharge rate is higher. Conversely, if the recharge rate of the aquifers decreases due to an arid to desertic climate shift in MENA regions, there could be a slight increase in concentration over a few years, owing to reduced dilution. The magnitude of the negative natural recharge shift would determine the extent of this increase.

We consider a general conceptual model (Fig. 11) of a contaminated site characterized by residual contamination (metallurgical and radioactive) in the vadose zone and groundwater in the study area. The contaminant plume from phosphate regions (M'Dilla, Métlaoui, Moularés, and Rédayef) migrates vertically through the vadose zone and laterally downgradient in the shallow multi-aquifer (sandy aquifer). At some sites, the plume reaches surface water bodies (Thelja and El Melah rivers) located close to the site boundary, through water seepage from phosphate washing and phosphogypsum valorization. To reduce contaminant migration through the vadose zone, the contaminant source zone (i.e., seepage basin) is often capped with low-permeability material, such as clay or silt. The model presented indicates that residual contaminants in the vadose zone, particularly at the final outlet of the watershed, have the potential to persist as a source of radioactivity and metallurgical contamination for the transboundary aquifer NWSAS (North Western Sahara Aquifer System) groundwater plume (increasing of H_2S (gas) by bacterial sulfur reduction, increasing ^{222}Ra (gas) by radiogenic disintegration…). Additionally, this could lead to atmospheric pollution and toxicity caused by radioactivity (Fig. 11).

Potential mitigation and Remediation strategies

Mitigation and remediation of groundwater impacted with dissolved metals, metalloids, and radionuclides is perhaps one of the biggest challenges for in situ environmental remediation today.

A- Phytoremediation

The processus of phytoremediation strategies can be summarized in 5 important steps in 2 places (surface and sub-surface areas) (Fig. 12):

1. absorption: from the saturation zone of shallow aquifer (contaminated zone);
2. rhizofiltration and rhizoaccumulation: contaminants are uptake by the roots of hydroponically grown plants, translocated, and accumulated into plant shoots and leaves. In the polluted area of the study area in the drainage network (Thelja, Magroun and El Maleh

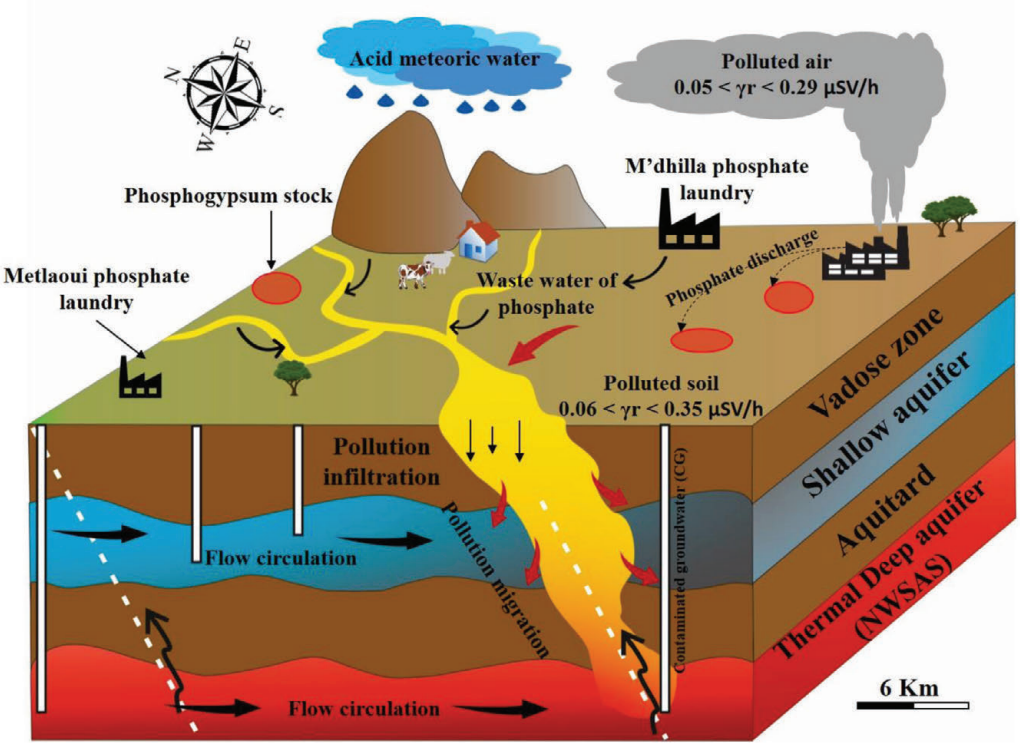

Fig. 11 - Conceptual model showing the paths of the migration of the contaminants in the study area.

Fig. 11 - Modello concettuale che mostra i percorsi di migrazione dei contaminanti nell'area di studio.

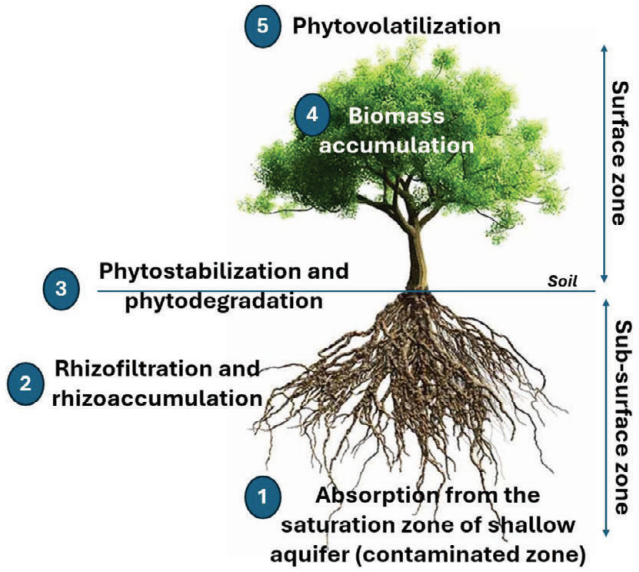

Fig. 12 - Phytoremediation processes adopted in the study area.

Fig. 12 - Processi di fitodepurazione adottati nell'area di studio.

rivers) and in the continental depression of El Gharssa Chott. Rhizofiltration has been demonstrated at several scales (bench-scale and field tests) effectively reducing the uranium concentration in phosphate wastewater. Rhizofiltration and rhizoaccumulation have been used to clean up waters contaminated with PTEs (Pb, Cd, Cu, Fe, Ni, Mn, Zn, Cr) and toxic radionuclides series (^{238}U, ^{234}Th and ^{222}Ra). In the Laboratory (LAM3E) scale rhizofiltration of tomato experiments: at a pH of 3, the ability to accumulate toxic radionuclides was 2 times higher than it was for solutions of pH 7 and pH 9 using TERRA in situ measurement;

3. phytostabilization and phytodegradation: These processus can be effected in the sub-surface and in the surface area of the plants ;
4. biomass accumulation: The green part of plants can accumulate a large quantity of RN and PTEs. But this accumulation depends on the variety of plants ;
5. phytovolatilization: During the photosynthesis process the gas can be volatilized to the large atmosphere.

B- Natural remediation

1. Aeration: volatilization from the high air evaporation (more than 1600 mm/year). This state encourages the aeration of gas RN (series of Rn-222) in the stagnation area of the industrial wastewater discharge of El Gharssa chott;
2. clay mineral: this mineral naturally exists in the phosphate discharge, it is characterized by high adsorption of PTEs and RN and after saturation, the clay level can inhibit the infiltration to saturated aquifer zone.

C- Other remediation methods

1. Physical and chemical methods: Micro, ultra, and nanofiltration processes work better to remove fine particles (0.001-0.1 microns). In the ultra-filtration separation, the contaminated liquid may need to be pretreated to form larger molecular complexes (e.g., metal-polymers or chelates) which are, then, more easily separated by the membranes. Membrane filtration technology may be considered when radionuclide and heavy metal contaminants are associated with suspended solids in a liquid media or when precipitating agents are available for pre-treating the liquid media (Benes et al., 1983; Chellam and Clifford, 2002). According to the bibliographic research (U.S. Environmental Protection Agency, 2001; Federal Remediation Technologies Roundtable, 2002), many other chemical methods are used in this type of RN, REEs and PTEs remediation but not at a large scale: granular activated carbon, membrane filtration technology, electro-remediation, physical separation technologie. In the large scale, these remediation methods need a high budget and a long time depending on the area, vadose zone lithology of contaminated area, wastewater composition, dispersion and the hydrodynamic of these contaminants ;
2. microbial methods: these methods have been developed to bioremediate areas contaminated with high uranium concentrations at low pH and high nitric acid concentrations (Wu et al., 2006). The different microbial-radionuclides and PTEs interaction mechanisms include biomineralization, biosorption, biotransformation and intracellular accumulation (Brookshaw et al., 2012). At a large scale, this bioremediation method also needs a high budget and a long time depending on the specificity of the study area, lithology and mineralogy variation of contaminated area, wastewater composition, dispersion and the hydrodynamic of the radio-toxic/PTEs contaminants;
3. specific membrane: in the study area, we adopt a specific membrane in the gas pipes to reduce the dust (GHGs) concentrations in the atmosphere;
4. increase the length of the gas channels: In the phosphate valorization stations (GCT) of phosphate transformation into fertilizers and sulfuric acid. To decrease the atmospheric particles (GHGs: green……) impact on soil, vegetation, human health…We adopt this method to create a dispersion or dilution of these particles by the winds (reduce the cumulative impact on air, water, soil and human health);
5. sea water transfer: Since its discovery, phosphate has been washed by brackish groundwater in the order of 5T of water/1T of phosphate. Starting from 2008, a strategic model has been proposed by Hamed et al. (2008). The model concerns the possibility of washing phosphate with seawater transported by pipelines from the Gulf of Gabes to the mining basin. This pilot project will boost the Tunisian phosphate economy, solve the problem of overexploitation of groundwater in the southwest of Tunisia, and will be located within the framework of respecting carbon, water and

environmental footprints.

The future outlook of this study is to pass to the CO_2 fluid injection technology and the monitoring wells (sensor real time of quanlity and quantity) in the polluted area to control the propagation of these toxic radionuclides and PTEs in the phosphate mining basin. The modeling is also required as well as the evaluation of radionuclide reduction rates, pathways, and prediction of the radionuclide concentration at the downgradient exposed receptor. One limitation for this study, except the artificial recharge (CO_2 fluid injection with the industrial wastewater), is that we can't effect all the remediation strategies because on a large scale, some of the physical, chemical or bioremediation methods need a high budget and a long time depending on the area, lithology variation, wastewater composition, dispersion and the hydrodynamic of the contaminants. The laboratory experiences in a small scale have been done. The phytoremediation has been done on a large scale along all the rivers and around the continental depression of phosphate wastewater discharge of El Gharssa Chott. All these remediation strategies have their advantages and limitations.

Radioactivity impact on human health

The unconventional uranium mining transboundary basin suffers from the disintegration of the natural and the anthropogenic radionuclides (^{238}U, ^{236}Th, ^{228}Ra, ^{226}Ra, ^{222}Rn and ^{222}Rn*) and the impact of the radio-toxic elements from the intrusive/sedimentary rocks, hydrodynamic of the hydrothermal reservoir of NWSAS and the atmospheric dust circulations on human health and on the environment are obvious and severe in the study area. In fact, the study area is characterized by a high rate of cancer (lung, breasts and prostate). In Gafsa/Tebessa phosphate mining basin has shown an increase in lung cancer rates in the region during the period (1997-2024) from 5 % to 15 %. This is due to the human exposition time (working inside caverns and living near the industrial phosphate sector) (Hamed et al., 2024a,b).

Measurements of radioactivity near phosphate-U levels in Tunisia's Gafsa basin showed a reading of 0.36 mSv/h. This demonstrated that air and soil radioactivity levels were elevated due to phosphate-U contamination in the Tunisian southern part (Gafsa/Tozeur/Kebili areas). In situ measurements of radioactivity rates (alpha and gamma) using a specific Terra-radiometer displayed variable values depending on the type of release (phosphate-U or PG wastewater). These negative impacts significantly affect the soil structure and texture and the quality of surface and groundwater aquifers.

Air and alimentation food in the agriculture lands (Gafsa, El Guettar, El Hamma, Tozeur, Nefta Oases) with toxic gas and particles propagation such as radionuclides (^{238}U, ^{234}Th, ^{224}Ra, ^{222}Rn, ^{222}Rn* and ^{40}K) and aerosols (particles dust, gas and PTEs…) caused many other health diseases (thyroid, breast and prostate cancers). The contamination variability depends on sex, age, working at CPG or not, life in the phosphate basin or not, working time (8h to 12h), exposition time (1 day to 365 days during around 75 years). Possible additional factors include smoking, alcoholism, psychologic and genetic parameters.

The dissolution of uraninite by the thermal groundwater of NWSAS (dissolution catalyst) during the residence time (0.2 m/y to 5 m/y using radiocarbon isotopes 13C/14C) to the transboundary Djerid/El Gharsa Chott area (paleo-lake) is shown in this equation:

Rock-$2UO_2$ (solid) + $4H^+$ (aq) + O_2 (gas) + H_2O (thermal water) $\rightarrow$ $2UO_2^{2+}$ (aq) + $2H_2O$ (aq)

Many other exchanges in groundwater can be effected between the toxic radionuclides and Ca, CO_3 and Pb integrated in the carbonate (calcite and dolomite) minerals.

Conclusion and recommendations

This study is derived from the investigation and assay of phosphate ore samples collected from the Gafsa mining basin, where the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K were calculated. Based on these measurements, the radiation indices consistently revealed values exceeding both recommended and permissible limits for all the phosphate samples and some hydrothermal groundwater samples. This includes a significant radioactivity level in the region. Radiation indices calculations are correlated to the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K with higher values than the recommended and permissible limits for these samples. The elevated results (between 0.31 and 0.36 mSV/h) are mainly due to the high contents of ^{238}U, ^{232}Th, ^{226}Ra, and radionuclides in the analyzed samples (gas, solid or liquid) that exceed IAEA guidelines (0.3 mSV/h).

The conducted surveys and sampling results demonstrate a significant hazard from radioactive materials impacting the environment and the human health in the study area. While some locations necessitate specific monitoring and remedial actions to mitigate associated risks, caution is advised against using these materials as building materials and their unrestricted use as agricultural amendments due to the substantial radiation hazard. Strict precautions are urged for workers in open-pit quarries exposed to high levels of radioactivity particularly the impact of Radon (toxic gas). Periodic monitoring of air, soil and water resources radioactivity is recommended to prevent unnecessary radiation exposure to the abiotic and biotic ecosystem. Additionally, measures to minimize and regulate the use of phosphate fertilizers, potential sources of radiation exposure to farmers and the general public are crucial. Implementation of remediation and content mitigation methods is deemed essential in this industrial phosphate mining basin. This icludes including the establishment of close protection zones to safeguard against urbanization in radioactive industrial areas. Furthermore, significant measures are vital for the residents' health in these mining regions to address any risk of contamination with a focus on early cancer control. Water and soil monitoring in real data is a necessity to avoid the deterioration of the quality of the water resources and the soil salinization.

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Author contributions
Younes Hamed: Methodology, Formal analysis, Data curation, Visualization, Writing-original draft preparation. Matteo Gentilucci and Hassen Jallouli: Conceptualization Methodology, Supervision, writing-review and editing. Kaouther Ncibi, Riheb Hadji and Abderraouf Jraba: Methodology, Formal analysis, Data curation, Visualization. Ammar Mlayeh and Elimame Elaloui: Conceptualization Methodology, Supervision, Writing-review and editing.
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