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Research Paper

PET plastics as a Trojan horse for radionuclides

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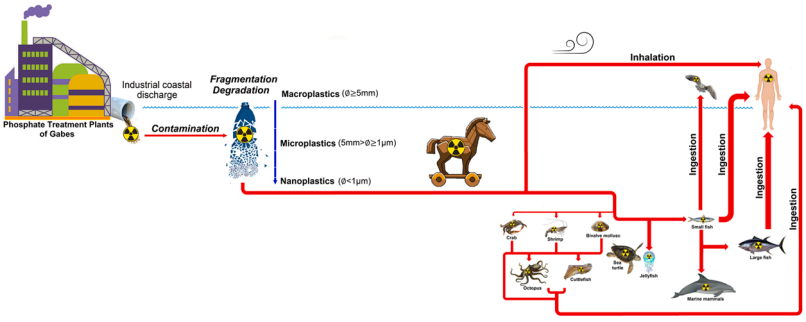
HIGHLIGHTS

- Radionuclides were characterized in PET bottles collected from coastal environment.
- Mean activity concentration of radionuclides associated with PET was 5.30 $Bq{\cdot}kg^{-1}$.
- Human health risk assessment indices were below international safety limits.
- PET can be loaded with natural and artificial radionuclides.
- PET can act as a carrier to transfer radionuclides to humans.

GRAPHICAL ABSTRACT



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ABSTRACT

Mismanaged plastic waste interacts with secondary environmental pollutants, potentially aggravating their impact on ecosystems and human health. Here we characterized the natural and artificial radionuclides in polyethylene terephthalate (PET) bottles collected from the industrial littoral discharge of a phosphate fertilizer plant. The activity concentrations in littered bottles ranged from 0.47 (^{208}Tl) to 12.70 $Bq{\cdot}kg^{-1}$ (^{226}Ra), with a mean value of 5.30 $Bq{\cdot}kg^{-1}$. All the human health risk assessment indices (annual intake, annual effective dose, and excess lifetime cancer risk) estimated for radionuclides associated with ingestion and inhalation of microplastics were below international safety limits. Our results demonstrated that PET can be loaded with natural and artificial radionuclides, and potentially act as a carrier to transfer radionuclides to humans, posing a new potential health risk. Increased use, mismanagement and fragmentation of plastic waste, and continued interaction of plastic waste with radioelements may lead to enhanced radiation exposure in the future.

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1. Introduction

Microplastics are ubiquitous in the environment: in the atmosphere (Allen et al., 2020), soils (Koutnik et al., 2021), sediments (Martin et al., 2020; Niu et al., 2021), seawater (Zayen et al., 2020), groundwater (Huang et al., 2021), lagoons (Wakkaf et al., 2020a, 2022), lakes (Mao et al., 2021), deserts (Wang et al., 2021), seafood (Smith et al., 2018), and even in the human placenta (Ragusa et al., 2021) and feces (Zhang et al., 2021). The contamination of the natural environment with plastic has been on the rise since the 1950s with the increasing dependence of almost all human activities on polymer materials. The worldwide mismanagement of plastic waste has made it a widespread and common pollutant (Geyer et al., 2017) and plastic waste presents global environmental, ecological, economic and health risks and challenges, as a result (Koelmans et al., 2022). A particular issue is the interaction of plastic waste with other contaminants. Recent studies have focused on microplastic particles and their ability to fix heavy metals (Gao et al., 2021), hydrocarbons (Song et al., 2021), pesticides (Li et al., 2021), pharmaceuticals (Puckowski et al., 2021), and pathogens (Pham et al., 2021). However, to our knowledge, no information is available in the literature about the radioactive contamination of plastic objects and their environmental and health repercussions.

A multitude of studies have been carried out to determine the level of natural and/or artificial radionuclides content in soils (Kang et al., 2021), sediments (Heldal et al., 2021), groundwater (Sherif and Sturchio, 2021), seawater (Byun et al., 2021), rivers (Guerrero et al., 2021), and foodstuffs (Haase et al., 2021). These various studies reported that the radioactive contamination of these compartments could constitute a potential risk for human health, mainly by ingestion, inhalation, and dermal contact (UNSCEAR, 2008). Two recent experimental studies have illustrated the sorption of radionuclides on commercial microplastics (Johansen et al., 2018; Ioannidis et al., 2022). Measuring the radioactivity of environmental microplastics would be challenging due to the substantial mass (grams) required. Therefore, we investigate in this study radionuclide interaction with plastic debris in the Gulf of Gabes littoral ecosystem, where phosphate fertilizer factories and urban runoff discharge a waste stream rich in radioactive compounds (phosphogypsum) and mismanaged plastics.

Gabes is a coastal industrial city located at the southeastern part of Tunisia (Fig. 1). It is considered as one of the most polluted Mediterranean regions, according to a study conducted by the Facility for Euro-Mediterranean Investment and Partnership and the World Bank (EIB, 2004; El Zrelli et al., 2018a). The industrialization of the Gabes coast started in the 1970s with the establishment of one of the largest industrial complexes specializing in the transformation of phosphate ores to fertilizers, currently named the Tunisian Chemical Group (or *Groupe Chimique Tunisien*; GCT). This was followed by the foundation of many other chemical industries (such as the Chemical Society Alkimia, Fluorine Chemical Industries (ICF), Timab Industries, PET Cristal Industries, OMV Tunisia, etc.). The inhabitants of Gabes city, especially those living in the vicinity of the GCT phosphate fertilizer factories, in Chatt Sidi Abd Essalam (Fig. 1) are exposed to high external radiation levels, in comparison with typical background values recorded in the literature (El Zrelli et al., 2019c). This radiological risk is directly related to the marine discharge of untreated phosphogypsum (PG) waste, the aerial dispersal of phosphate ores stored at factories (El Zrelli et al., 2019c) and accumulated on the beaches after its marine discharge (El Zrelli et al., 2019a), and the phosphogypsum foams which are formed in seawater after discharging the PG waste (El Zrelli et al., 2019b). It is estimated that ~30 × 10^3 t of humid PG (14,4 ×10^2 t of dry PG; El Zrelli et al., 2017) are discharged daily in the marine environment of the Gulf of Gabes, so that the accumulated waste exceeds 200 × 10^6 t of dry PG, since the establishment of the local phosphate industry, in 1972 (El Zrelli et al., 2018b). The contamination of the marine environment in the Gulf of Gabes with various industrial pollutants, including phosphogypsum-derived radionuclides, was reported to be responsible for the degradation of local habitats and biodiversity (Darmoul et al., 1980; Darmoul, 1988; Pergent and Kempf, 1993; El Zrelli et al., 2015; El Zrelli, 2017; El Kateb et al., 2018; Hamza et al., 2021) and for health impacts on both marine life and humans (El Zrelli et al., 2019c, 2020, 2021; Rabaoui et al., 2014, 2015, 2017; El Kateb et al., 2016).

Human populations are constantly exposed to external (e.g., from terrestrial radiation and cosmic rays) and internal radiation (through the ingestion and inhalation of natural and artificial radionuclides) (Basu et al., 2015). Natural radionuclides, in particular the decay chains of ^{238}U and ^{232}Th are ubiquitous in the environment, whereas artificial radionuclides, associated with release from the nuclear weapon and energy industries, are less common. Recent studies reported that

Fig. 1. Location of the study area.

microplastics are present in food webs, drinking water and air, and that humans are exposed to microplastics via dietary intake and inhalation (Cox et al., 2019; Nor et al., 2021; Senathirajah et al., 2021). Since microplastics can be loaded with many harmful pollutants (Issac and Kandasubramanian, 2021), potentially including radionuclides, it is probable that the ingestion and inhalation of radionuclide-loaded microplastics exacerbate the exposure of humans to radiation. To our knowledge, there is a lack of information on the levels of radioactivity in plastics and on the potential human health hazards related to the ingestion of radioactive plastics. The present work was conducted in this regard and aims at *i*) characterizing the capacity of polyethylene terephthalate (PET) plastics collected from the human-impacted coastal area of Gabes Gulf to host environmental radionuclides, and *ii*) assessing the potential impacts of ingestion and inhalation of radioactive plastics on human health in the area surrounding the industrial complex of Gabes. The human health risk assessment includes the estimation of annual dietary intake, annual effective dose, and excess lifetime cancer risk associated with the direct ingestion of PET particles.

2. Materials and methods

2.1. Sampling and samples' preparation

To achieve the goal of this study, 43 discarded PET bottles used for mineral drinking water packaging and thrown in the natural environment were collected from the contaminated coastal area of the Gulf of Gabes (close to the GCT industrial complex of Gabes; PET_C). Only PET bottles with closed caps were selected for this study and were collected from the edges of the littoral industrial discharge mouth of the GCT complex (Fig. 2). To compare the characterization of the surface PET_C samples with those of non-used PET, 24 virgin drinking water bottles (PET_V) were additionally collected from local markets. All sampling was carried out in August 2020. The choice of PET bottles was not arbitrary because they are the dominant consumed plastic product for drinking water by the local population (due to the deteriorated quality of tap water), they are non-reusable, and they represent the most abundantly observed macroplastic debris in the GCT industrial littoral discharge (Fig. 2C). Furthermore, PET bottles are not recycled in Gabes, and "illegal" dumping in the valley and fishing harbor of the city is a common practice, as shown in Fig. 2A and B. Therefore, the heavy daily use of PET bottles and the mismanagement of their waste make PET bottles the most polluting plastic material in Gabes (Fig. 2).

To remove any surface contamination and adsorbed impurities (shells, sediment particles, aquatic organisms, dusts), all PET bottles (PET_V and PET_C) were briefly rinsed with ultrapure water following established protocols (Milli-Q®, at 25 °C; Kedzierski et al., 2018). The bottle samples were dried in a hot air oven (24 h at 25 °C) and then cut into small pieces of 3–5 mm^2 using a pair of ceramic blade scissors. The two groups of PET pieces were separately homogenized and stored in an airtight glass container prior to radiological analysis.

Fig. 2. Mismanaged waste accumulation of PET bottles in the valley (A), fishing harbor (B) and edges of the industrial littoral discharge mouth of phosphate treatment plants of Gabes (C).

2.2. PET bottle surface characterization

The surface topography and roughness of both PET_V and PET_C bottles were characterized using Focused-Ion-Beam/Scanning Electron Microscopy (FIB/SEM; Helios NanoLab™ 600i, Thermo Fisher Scientific) coupled with a Silicon Drift Detector Energy Dispersive Spectrometer (X-Max® 80 mm^2 SDD-EDS; Oxford Instruments) at the Raimond CASTAING Micro-characterization Center (Toulouse, France). The Leica sputter coater EM ACE600 was used to coat the sample surfaces with a layer of carbon necessary for electron microscopy images and analysis. Two types of image capture modes were used: secondary (SEI; for surface topography) and backscattered (BSE; for chemical contrast) Electron Imaging modes. The thickness of the contaminants layer on the surface of PET_C was determined by xT Microscope Control software and its chemical composition by AZTec X-ray microanalysis software.

2.3. Analytical procedure

Three naturally occurring series of radioactive elements exist. The parent nuclides are ^{238}U, ^{235}U and ^{232}Th that generate series of radionuclides - with different half-lives and chemical behaviors - following alpha or beta decay. Several daughters of these series are also gamma emitters and will be determined in the present study using gamma spectrometry. In natural samples, the daughter nuclides of the series may equal the activity of their parent nuclides (radioactive secular equilibrium), as is the case in closed systems. Alternatively, radioactive disequilibrium may also occur in the decay series, as a consequence of the migration of one daughter nuclide due to physical or chemical processes. The samples were dried, weighed (PET_V=41.39 g; PET_C= 32.75 g) and placed in sealed containers (72 ×15 mm) to prevent any radon gas loss (^{222}Rn being part of the ^{238}U decay series). Prior to analysis, the samples were stored for 3 weeks which are necessary to reach radioactive equilibrium between U, Th, and their short-lived decay products, in case ^{222}Rn escaped during the sample preparation. The analysis of samples was made using low-background gamma spectrometry at the LAFARA underground laboratory in the French Pyrénées (van Beek et al., 2013). The gamma spectrometers at the LAFARA are placed under 85 m of rock, which protect the detectors from cosmic radiations, thus providing a very low background to the counting facility. An ORTEC-AMETEK semi-planar high purity germanium (HPGe) detector of 183 cc (van Beek et al., 2013) equipped with a CP5®+ electric cryogenerator and LYNX® electronics (MIRION-CANBERRA) was used. Each individual sample was analysed during 96 h. Data acquisition and treatment of the spectra were performed using APEX software (MIRION-CANBERRA). The spectrometer calibration has been performed using IAEA reference materials RGU-1 (^{238}U and daughters at secular equilibrium), RGTh-1 (^{232}Th and daughters at secular equilibrium) and IAEA-447 (^{40}K and ^{137}Cs).

The activities of gamma emitters from the decay chains of ^{238}U (^{234}Th, ^{226}Ra and ^{210}Pb) and ^{232}Th (^{228}Ra, ^{228}Th and ^{208}Tl), as well as ^{40}K and ^{137}Cs were determined. From the ^{238}U decay chain, we determined the activity of ^{234}Th (63.3 keV), ^{226}Ra (using the γ rays of ^{214}Pb at 295.2 keV, 351.9 keV and ^{214}Bi at 609.3 keV) and ^{210}Pb (46.5 keV). The samples were analyzed after more than six months from the collection date and given the short half-life of ^{234}Th (24.1 days), the ^{234}Th activity is considered in equilibrium with its direct parent radionuclide ^{238}U (i.e., no remaining excess of ^{234}Th in the samples). The analysis of ^{234}Th thus provides a quantification of the ^{238}U activity in the samples. From the ^{232}Th decay chain, we determined the activity of ^{228}Ra (using the ^{228}Ac γ rays at 338.3, 911.2 and 969.0 keV, with ^{228}Ac assumed to be in equilibrium with ^{228}Ra), ^{228}Th (using the γ rays of ^{212}Pb at 238.6 keV), and ^{208}Tl (using both γ rays at 583.2 and 2614.5 keV). Note that within the ^{232}Th decay chain at secular equilibrium, the ^{208}Tl activity is expected to be ca. 1/3 of the ^{228}Ra (or ^{228}Th) activity due to the specific branching scheme in the decay chain ahead of ^{208}Tl. Finally, we reported activities of ^{40}K (1460.8 keV), as well as activities of the artificial radionuclide ^{137}Cs (661.7 keV), when this latter was detected in the samples. The detection limits are 0.96, 0.26, 1.33, 0.39, 0.16, 0.09, 2.48 and 0.10 Bq kg^{-1} for ^{234}Th, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{40}K and ^{137}Cs, respectively.

2.4. Assessment of human health risk

The probable effect of the ingestion and inhalation of radionuclide-loaded PET microplastics on human health of local inhabitants was assessed using three indices: the Annual Intake (AI), Annual Effective Dose (AED), and Excess Lifetime Cancer Risk (ELCR). We made the important assumption that the plastic debris we observed contribute to local microplastic exposure via the consumption of seafood and via inhalation of MPs in air.

2.4.1. Annual Intake

The Annual Intake through the direct ingestion and inhalation of radionuclide-loaded PET microplastics was estimated for both PET_V and PET_C samples using the following equation:

$$AI_T = \sum_{i=1}^{n} AI = \sum_{i=1}^{n} 2C \times A \quad (1)$$

with:

AI_T: sum of individual annual intake for all analyzed radionuclides.

AI: individual annual intake for each analyzed radionuclide.

C: activities (in Bq kg^{-1}) for each analyzed radionuclide, multiplied by 2 because the debris analyzed (closed bottles) only hosted radionuclides on half of its available surface area.

A: annual plastic rate consumption (in Kg y^{-1}) per person. Table 1 lists human microplastic exposure estimates from the literature in particles per capita per year (#/capita/y) and μg per capita per year (μg/capita/y) for dietary intake and inhalation. Particle to mass conversion is critical in assessing radiation exposure here because radioactivity is measured per mass unit (in Bq kg^{-1}). Exposure data from Senathirajah et al. (2021) showed inexplicably high mass exposure (from 0.1 to

Table 1
Adult human intake of microplastics, including dietary ingestion and inhalation. Seafood includes fish and shellfish. MP: microplastic.

Plastic intake based on:	Population		MP intake		References
			#/capita/y	μg/capita/y	
shellfish, salt, drinking water, and beer	Global Global	min max	102,527 102,527	770×10^4 287×10^6	Senathirajah et al. (2021)
seafood, salt, drinking tap water, alcohol, and honey (15 % of caloric intake)	USA	mean	44,414	251	Cox et al. (2019)
Inhalation	USA	mean	46,440	18	
Total intake: sea food, drinking water, salt, beer, milk, and air inhalation	Global	median	322,295	213	Nor et al. (2021)
Inhalation	Global	median	98,237	39	
Ingestion (15 % caloric correction)	Gabes (Tunisia)	mean	1,222,363	1546	*Present study*
Inhalation			72,339	29	
Total intake			1,294,702	1575	

5 g/week), given similar estimates of particles exposure as alternative studies. Cox et al. (2019) provided both dietary and inhalation particles exposure estimates, which we converted to mass exposure using the mean microplastic particle masses from Nor et al. (2021) which are equal to 5.65×10^{-6} and 3.97×10^{-7} µg/particle for food and air, respectively.

2.4.2. Annual effective dose

The 'Annual Effective Dose' index (UNSCEAR, 2000) has been found useful in the assessment of the radiological risk because it gives a good idea on the total dose proportion received by a particular population (Makon et al., 2011; Uwatse et al., 2015). The AED can be calculated as follows:

$$AED_T = \sum_{i=1}^{n} AED = \sum_{i=1}^{n} AI \times e \quad (2)$$

with:

AED_T: sum of individual annual effective doses for all analyzed radionuclides.

AED: annual effective dose for each analyzed radionuclide.

AI: individual annual intake for each analyzed radionuclide.

e: dose coefficient for ingestion and inhalation of each analyzed radionuclide (in Sv Bq^{-1}; ICRP, 2012; Delacroix et al., 2006).

2.4.3. Excess lifetime cancer risk

According to Qureshi et al. (2014), the ELCR can be calculated as follows:

$$ELCR = AED \times DL \times RF \quad (3)$$

with:

$ELCR$: Excess lifetime cancer risk.

AED: Annual Effective Dose.

DL: average lifetime duration, which is 76.70 years for Tunisian people in 2019 (WB, 2022).

RF: fatal risk factor per Sievert (0.05 Sv^{-1}; ICRP, 2008).

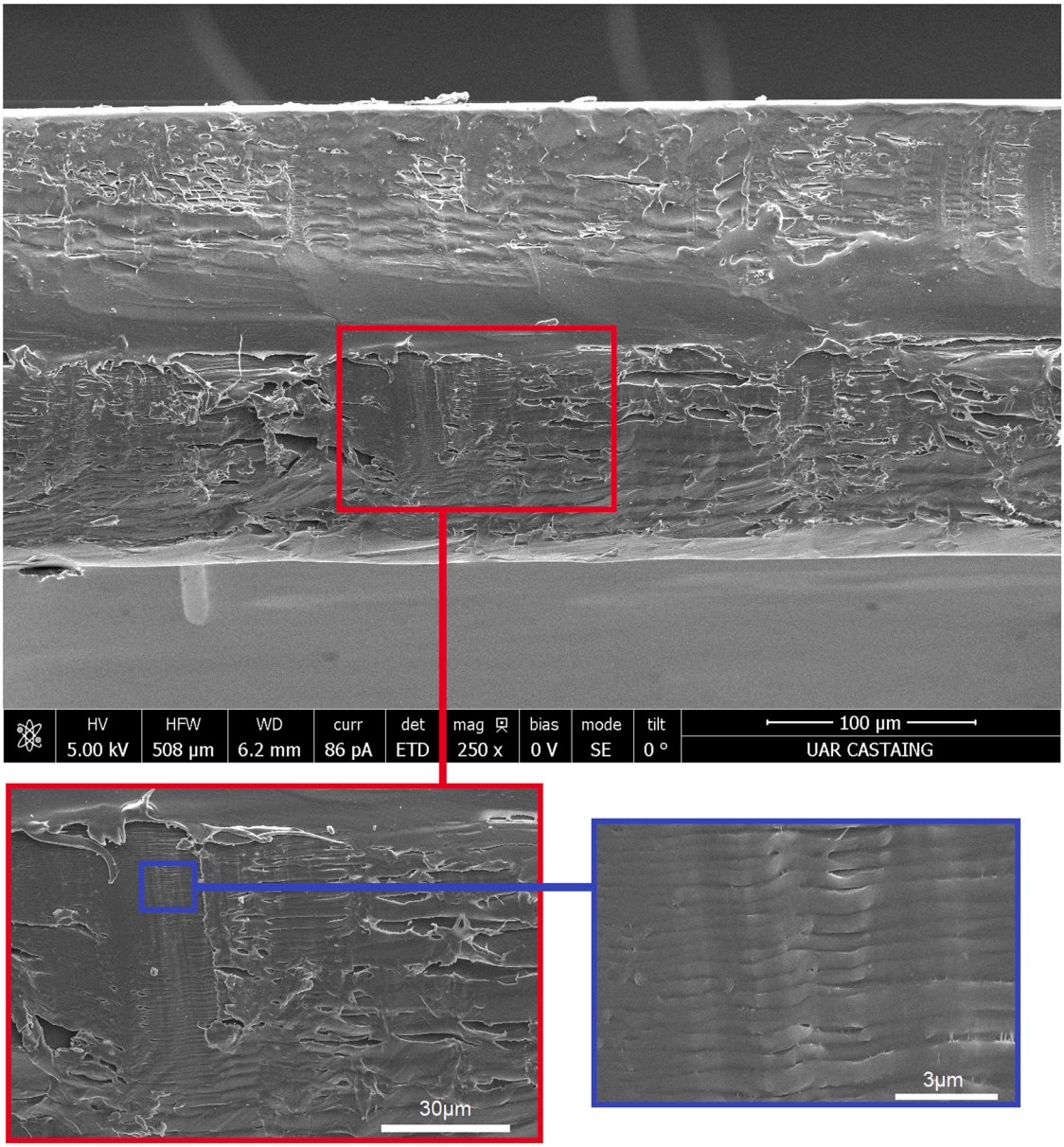

Fig. 3. The multi-layer structure of PET bottles.

3. Results

The FIB/SEM characterization showed that PET bottles are formed by several thin layers joined together (Fig. 3). Additionally, we noticed an evident morphological differences between the surfaces of virgin PET_V and contaminated PET_C bottles (Fig. 4). While PET_V bottles presented a smooth and homogenous surface (Fig. 4A), PET_C bottles were found to be rough and heterogeneous (Fig. 4B). The electron photomicrograph of a PET_C bottle longitudinal cut, given in Fig. 5, showed a brown to blackish and 6–19 μm thick layer (average thickness of 13 μm) which partially covers the PET surface (average PET thickness =214 μm). The examination of this superficial layer showed that it is mainly formed by contaminated organic matter, and phosphogypsum and marine halite crystals (Fig. 6). No microorganism colonies were observed at the surfaces of either PET_V or PET_C bottles.

Table 2 presents activity concentrations of ^{234}Th, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{40}K, and ^{137}Cs measured in PET_V and PET_C samples. These activity concentrations ranged from < 0.10 (^{137}Cs) to < 2.48 Bq kg^{-1} (^{40}K), and < 0.12 (^{137}Cs) to 12.70 Bq kg^{-1} (^{226}Ra), for PET_V and PET_C samples, respectively. The decreasing order of activity concentrations for PET_C was found to be ^{226}Ra $>^{234}$Th $>^{210}$Pb $>^{40}$K $>^{228}$Th $>^{228}$Ra $>^{208}$Tl $>^{137}$Cs. The activity concentrations of the radionuclides associated with the ^{232}Th decay chain (^{228}Ra, ^{228}Th, and ^{208}Tl) suggest that the radionuclides are at secular equilibrium in PET_C, the ^{208}Tl activity being lower due to the 35.6 % branching in the decay chain. The ^{228}Ra and ^{228}Th activity concentrations thus provide an estimate of the ^{232}Th activity concentration in the sample (1.2 Bq kg^{-1}). Similarly, the ^{234}Th activity concentration analyzed in the sample can be related to ^{238}U (8.8 Bq kg^{-1}) and we found that the daughters from the ^{238}U decay chain are in secular equilibrium, with a slight excess of ^{226}Ra. The ^{228}Ra/^{234}Th activity ratio reported in the PET_C is thus indicative of the ^{232}Th/^{238}U ratio (i.e., 0.1). We reported significant ^{40}K activity concentrations in the PET_C. The radionuclides from the ^{235}U decay chain are also expected to be present in the samples. However, because of the lower natural abundance of ^{235}U, these radionuclides are likely to have a negligible impact and were therefore excluded from further analyses. Finally, note that a peak of ^{137}Cs was detected in the PET_C sample but the activity concentration was below the detection limit.

Table 4 presents the estimated values of annual intake (AI), annual effective dose (AED), and excess lifetime cancer risk (ELCR) indexes (see Materials and methods), assessed based on the potential adult human intake (by ingestion and inhalation) of PET_C microparticles, contaminated with radionuclides, by local residents. Considering a microplastic ingestion and inhalation rate of 1.6×10^{-6} kg y^{-1} (Table 1), AI was found to range between 9.19×10^{-6} Bq y^{-1} (by inhalation) and 4.98×10^{-4} Bq y^{-1} (by ingestion), with a total annual effective dose of 5.07×10^{-4} Bq y^{-1}.

Similarly, the 'AED' index, estimated based on an annual rate of microplastic consumption equal to 1.6×10^{-6} kg, varied from 6.01×10^{-11} Sv y^{-1} (by inhalation) to 8.63×10^{-11} Sv y^{-1} (by ingestion), with an annual effective dose of 1.46×10^{-10} Sv for PET_C (Table 4). The AED value for exposure to PET_C is lower than the standard exposure values recommended for the public of 1 mSv y^{-1} (Akhter et al., 2007; Table 4).

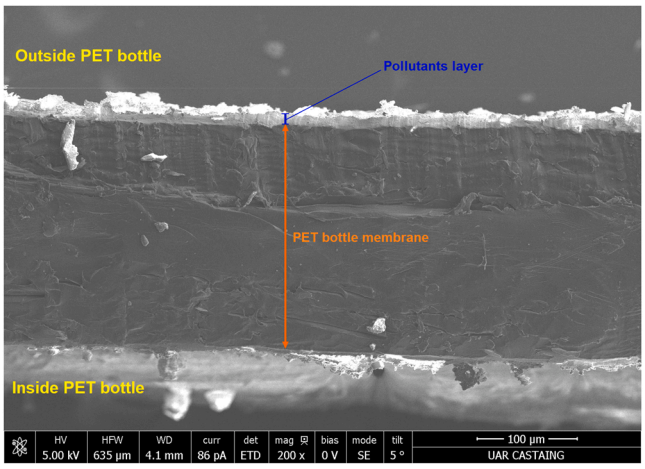

Fig. 5. FESEM image of a longitudinal cut of contaminated PET bottle (scale bar: 100 μm).

Regarding ELCR, the cumulative value of this index estimated for the local population (for a lifetime of 76.7 years), using the estimated microplastic ingestion rates of 1.6×10^{-6} kg, was 5.62×10^{-10} for PET_C (Table 4). Similar to AED results, all ELCR calculated values (ingestion and inhalation) were found to be below the cancer risk factor suggested by ICRP (Akhter et al., 2007; i.e., 2.5×10^{-3} based on an annual dose limit of 1 mSv for the general population).

Based on the total microplastic exposure estimated by both Cox et al. (2019) and Nor et al. (2021) (Table 1, dominated by seafood consumption and inhalation which are both relevant in the context of Gabes), we estimated a potential human microplastic intake of 1575 μg/capita/y (A=1.6 $\times 10^{-6}$ kg/capita/y in Eq. 1). Multiplied by the sum of the individual PET_C radionuclide activity concentration, and corrected for caloric intake and PET_C surface area, we found an AI of 5.07×10^{-4} Bq y^{-1}, (by ingestion and inhalation), an AED of 1.46×10^{-10} Sv y^{-1}, and an ELCR of 5.62×10^{-10} (Table 4).

4. Discussion

In recent years, PET plastic pollution studies have been greatly increasing and several researchers evidenced the ability of PET micro- and nano-particles (MPs and NPs) to adsorb different types of chemical and organic pollutants from their environment (Gao et al., 2021;

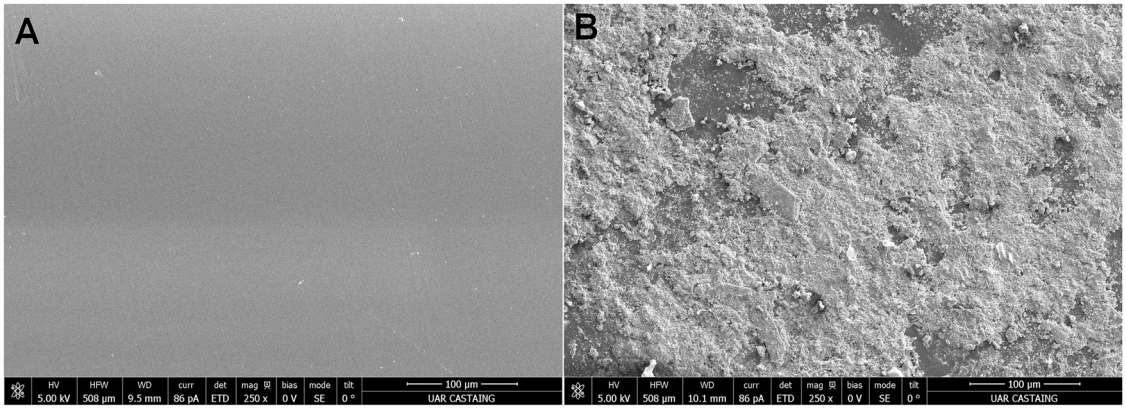

Fig. 4. FESEM images of the surface topography of virgin (PET_V; A) and contaminated (PET_C; B) PET bottles (scale bar: 100 μm).

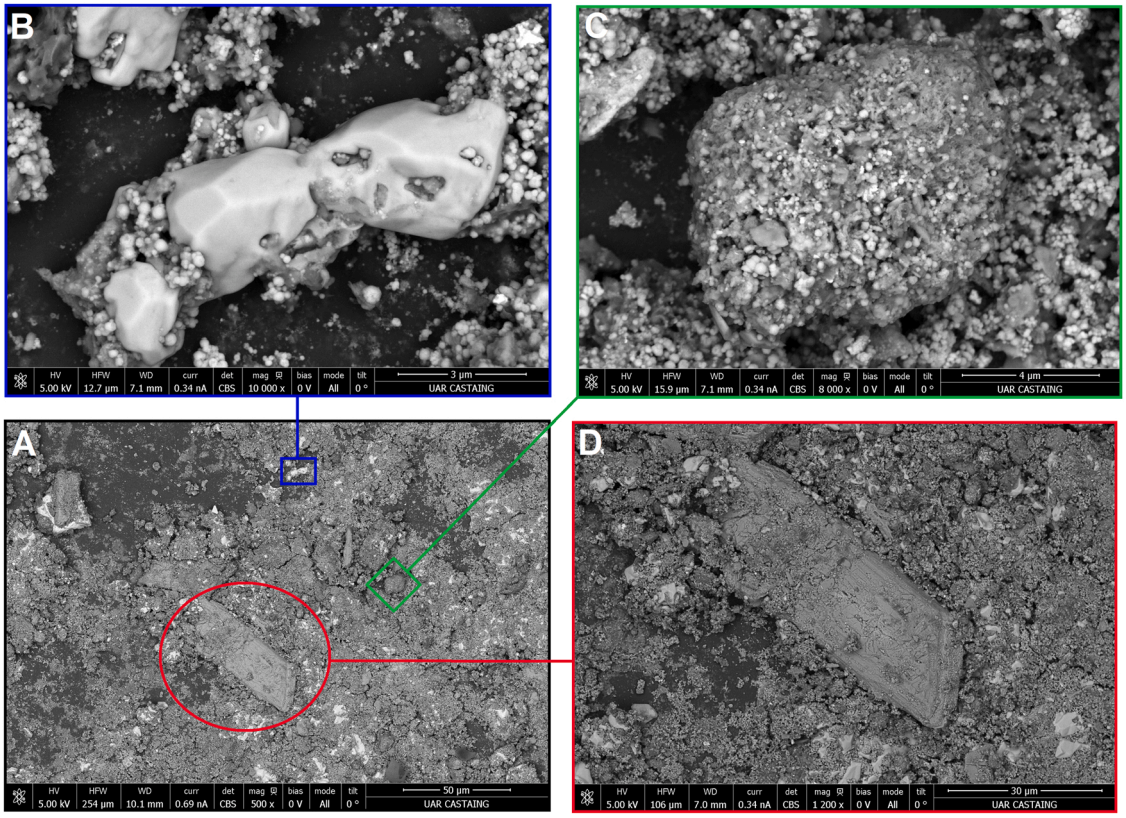

Fig. 6. FESEM images of a contaminant layer on the surface of a contaminated PET bottle (A; scale bar: 50 μm), halite (B; scale bar: 3 μm), contaminated organic matter (C; scale bar: 4 μm), and phosphogypsum (D; scale bar: 30 μm).

Table 2
Activity concentrations (in Bq kg^{-1}) of ^{234}Th, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{40}K, and ^{137}Cs in contaminated (PET_C) and virgin (PET_V) polyethylene terephthalate (PET) bottles. For PET_V, the measurement incertitude shown represent two-sigma uncertainty.

	^{234}Th	^{226}Ra	^{210}Pb	^{228}Ra	^{228}Th	^{208}Tl	^{40}K	^{137}Cs
PET_C	8.81 ± 1.83	12.7 ± 0.4	8.14 ± 1.71	1.17 ± 0.33	1.18 ± 0.17	0.47 ± 0.11	4.60 ± 2.43	<0.12
PET_V	<0.96	<0.26	<1.33	<0.39	<0.16	<0.09	<2.48	<0.10

Rochman et al., 2014; Lionetto and Corcione, 2021). In this work, and for the first time in the context of natural plastic debris, we showed the ability of these emerging environmental contaminants to carry natural and artificial radionuclides in urban-industrial discharges.

Due to the effects of various environmental factors including the wave forces, exposure to sunlight, and erosion by sand grains, the outer surface layers of the bottles (Fig. 3) cannot resist and gradually begin to degrade (Fig. 7). The degraded zones make the bottle surface rough, increase surface area, and therefore enhance the fixation of phosphogypsum foams (PGF) which are abundant in the industrial coastal discharge. These industrial foams adhere to the bottle surface by physical adsorption. The mechanism of radionuclide's sorption on the surface PET bottles is likely to occur in two essential steps: (*i*) degradation of PET bottle surface, and (*ii*) physical adsorption of PGF on the surface of PET plastics (Fig. 8).

Among all analyzed radionuclides, ^{226}Ra was the radionuclide that has the highest measured activity concentration (12.7 ± 0.4 Bq kg^{-1}; Table 2). This trend is similar to what was measured in phosphate rock (PR; ^{226}Ra=375.1 ± 1 Bq kg^{-1}), phosphogypsum (PG; ^{226}Ra=220 ± 1 Bq kg^{-1}) and phosphogypsum foams (PGF; ^{226}Ra=1169 ± 3 Bq kg^{-1}) which have been identified as the main source of radioactive pollution in Gabes marine environment (El Zrelli et al., 2019c). In addition, according to Table 3, the correlation between activity concentrations in PET, and PR (0.93), PG (0.78) and PGF (0.95) is positive and significant.

The mineralogical composition of the thin adsorbed layer on the surface of PET_C bottles (Fig. 4) was very similar to that of phosphogypsum foams with organic contaminated cement, phosphogypsum crystals, and other secondary minerals (*e.g.*, halite, quartz, sphalerite-Cd, fluorapatite, etc.) which are present in micrometric and nanometric forms. Note that the $^{232}Th/^{238}U$ ratio - derived from the $^{228}Ra/^{234}Th$ activity ratio - was 0.1, which is in good agreement with the $^{232}Th/^{238}U$ ratio determined in phosphogypsum foams (El Zrelli et al., 2019c). However, while the structure of phosphogypsum foams is alveolar, the PET_C surface deposits are in the form of clusters (Fig. 4B). The superficial alteration of PET_C bottles (appearance of thin scratches; Fig. 7) seems to play an important role in the fixation of this layer on the surface of bottles. In fact, the surface roughness of MPs is likely to facilitate the formation of deposits, compared to those with smooth surfaces (Yang et al., 2014; Fotopoulou, and Karapanagioti, 2015). These superficial morphological changes may be due to several abiotic factors such as physical abrasion and fragmentation in the discharge zone, by beach sand grains and wave action, but also by chemical corrosion due to the acidic pH of the GCT industrial effluents (pH = 3.11; El Zrelli et al., 2018a). Under the current chemical pollution conditions of the GCT industrial discharge, we consider it unlikely that biotic factors (bacteria, fungi, and algae) contribute to PET plastic degradation, as it has been shown in other studies (Moog et al., 2019; Sarkhel et al., 2020).

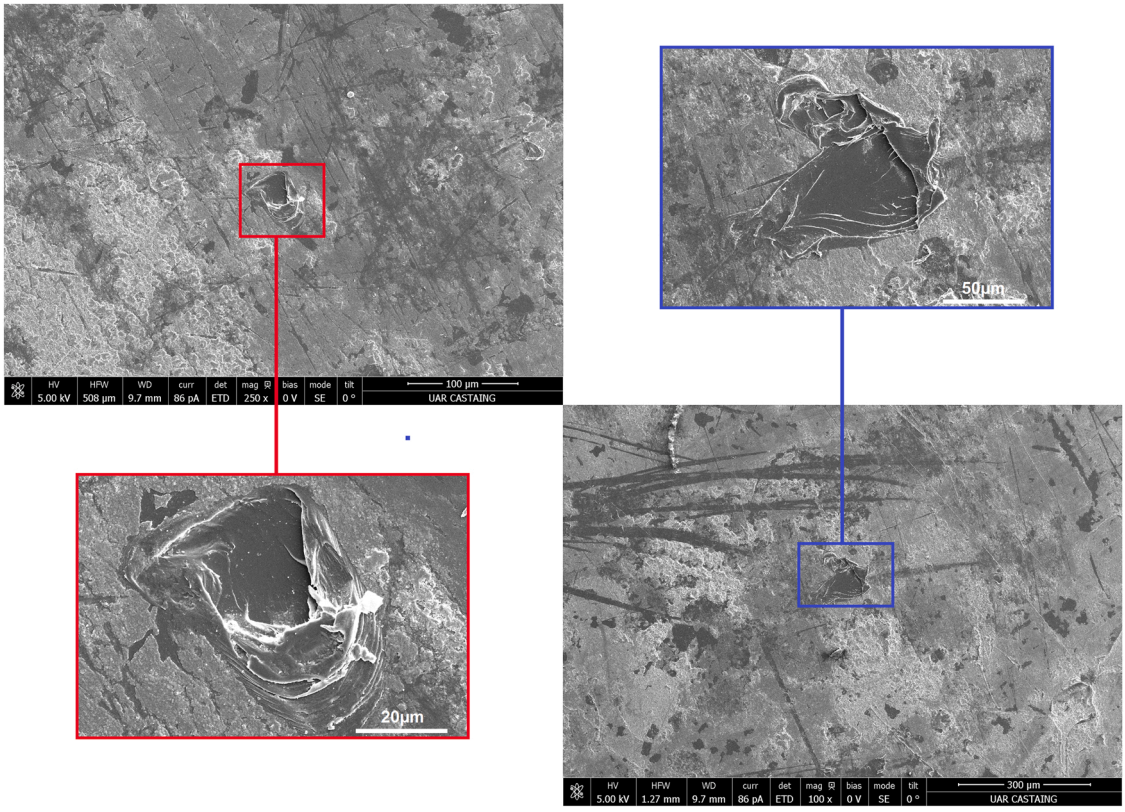

Fig. 7. Surface degradation of PET bottles.

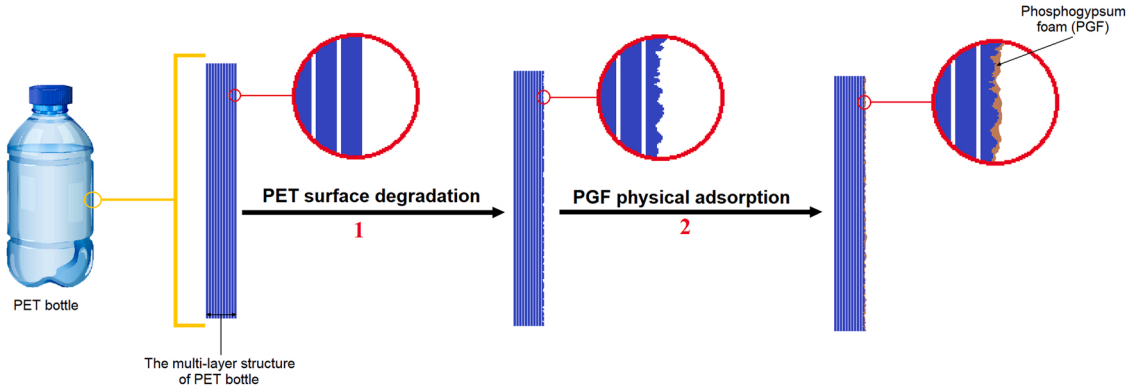

Fig. 8. Mechanisms of physical adsorption of phosphogypsum foams (PGF) on the surface of PET bottles.

Table 3
Correlation between the levels of radioactivity in PET (this study) and those in phosphate rock (PR), phosphogypsum (PG), and phosphogypsum foam (PGF; El Zrelli et al., 2019c).

	PET	PR	PG	PGF
PET	1.00	0.93	0.78	0.95
PR		1.00	0.76	0.54
PG			1.00	0.13
PGF				1.00

Table 4
Annual Intake (in Bq y^{-1}), Annual Effective Dose (in Sv y^{-1}), and Excess Lifetime Cancer Risk from ingestion and inhalation of contaminated polyethylene terephthalate microplastics (PET_C).

	Annual Intake (Bq y^{-1})	Annual Effective Dose (Sv y^{-1})	Excess Lifetime Cancer Risk
Ingestion	4.98×10^{-4}	8.63×10^{-11}	3.31×10^{-10}
Inhalation	9.19×10^{-6}	6.01×10^{-11}	2.31×10^{-10}
Total exposition	5.07×10^{-4}	1.46×10^{-10}	5.62×10^{-10}

In the broader littoral zone, several abiotic and biotic factors including ultraviolet and thermal radiations, wave action, oxidation, pH, salinity, biofilm formation, and microbial degradation could influence the degradation and fragmentation of the radionuclide contaminated macroplastic debris (∅≥5 mm) we observed, to microplastic particles (5 mm>∅≥1 µm) and to nano-plastics (∅<1 µm; Lionetto and Corcione, 2021; Wakkaf et al., 2020b; Atugoda et al., 2021). These mechanical, chemical and/or biological degradations/fragmentations increase MPs specific surface area (Fotopoulou and Karapanagioti, 2015) and hydrophobicity, which in turn increases their chemical

reactivity towards environmental pollutants (Atugoda et al., 2021; Joo et al., 2021) and hence their pollutant loads. More importantly, the increase in the pollutant loading of MPs can aggravate their potential ecotoxicological effects including on humans. In our study, we assumed that the radioactive plastic debris we observed can be degraded to microplastics in the coastal marine environment, where they integrate food webs, and can be emitted to the atmosphere (Allen et al., 2020). Human microplastic exposure is dominated by seafood consumption and inhalation of microplastics in air (Cox et al., 2019; Nor et al., 2021), which we took into account in our exposure estimate (see Materials and methods).

Even though the human health risk indices (AI, AED, and ELCR) did not exceed international safety limits, the radio-chemically contaminated PET_C debris should be considered as hazardous. Plastic debris in areas without enhanced industrial or natural radionuclide sources do not likely present measurable radioactivity. Continued and increased use of plastic waste, its mismanagement, and continued fragmentation of plastic debris in the Gulf of Gabes, and elsewhere, may lead to enhanced exposure in the future.

5. Conclusion and recommendations

We showed for the first time that it is possible for polyethylene terephthalate plastic debris to be contaminated by natural and artificial radionuclides from non-nuclear waste discharge, such as the phosphogypsum of phosphate fertilizer factories. We considered that fragmentation of radionuclide contaminated PET in the Gulf of Gabes likely leads to the contamination of seafood that is consumed by coastal human populations, and to emission of marine PET that can be inhaled. We quantified the Annual Intake, Annual Effective Dose, and Excess Lifetime Cancer Risks from ingestion of radionuclides contained in microplastics in a typical human diet in Gabes. Although we found that PET radionuclide exposure would be 7 times enhanced compared to virgin PET exposure, the ELCR remained below international dose limits. In parallel, international ICRP dose limits for radioelement exposure have been gradually lowered over the past decades, leading to a point where radioactive microplastics caused by mixed phosphate industrial and urban waste disposal could become a significant burden for human radiation exposure.

Better waste management of both phosphogypsum and plastic discharge is needed to avoid and/or reduce the potential fertilizer industry radioactive impact on the study area and elsewhere across the Mediterranean coast. Otherwise, the extensive usage of plastic, its short service life, its long (bio) degradation time, and its mismanagement will likely continue complicating the current environmental/health situation of Gabes and other similar littoral areas in the world. Finally, the radiological risk associated with micro/nano-plastic particle ingestion should be considered in future radiological and epidemiological studies. To precisely evaluate these radiological, environmental, and health effects, it is recommended to take into consideration all major routes of human incorporation of these emerging pollutants, especially by ingestion and inhalation.

Environmental implication

Although several studies have been conducted on microplastics as vector of organic and inorganic pollutants, only few studies were conducted so far on the -in vitro- association of radionuclides with microplastics. This is the first study confirming the association of natural and artificial radionuclides with polyethylene terephthalate bottles collected from the surrounding environment (from the industrial littoral discharge of a phosphate fertilizer plant). In addition to the determination of the activity concentrations of radionuclides in littered bottles, the study also assessed the human health risks associated to the inhalation and ingestion of radioactive PET microplastics.

CRediT authorship contribution statement

R.E.Z. and **L.J.R.** designed the study. **R.E.Z., L.Y., C.J., P.v.B., T.Z.**, and **M.S.** collected and/or analyzed all samples. **R.E.Z.** and **J.E.S.** calculated the indexes. **R.E.Z., L.Y., S.C, M.G., C.J., J.F.O., P.C.R., J.E.S., P.v.B, T.Z.,** and **L.J.R.** analyzed the data and interpreted the results. **R.E.Z.** wrote the manuscript with the support from all co-authors. **R.E.Z, S.C.** and **M.G.** provided the funding to complete the analysis. **R.E.Z., P.v.B., J.E.S.** and **L.J.R.** checked the manuscript before submission. **R.E.Z., J.E.S.**, and **L.J.R.** supervised the project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.129886.

References

Akhter, P., Rahman, K., Orfi, S.D., Ahmad, N., 2007. Radiological impact of dietary intakes of naturally occurring radionuclides on Pakistani adults. Food Chem. Toxicol. 45 (2), 272–277. https://doi.org/10.1016/j.fct.2006.08.006.

Allen, S., Allen, D., Moss, K., Le Roux, G., Phoenix, V.R., Sonke, J.E., 2020. Examination of the ocean as a source for atmospheric microplastics. PLoS One 15 (5), e0232746. https://doi.org/10.1371/journal.pone.0232746.

Atugoda, T., Vithanage, M., Wijesekara, H., Bolan, N., Sarmah, A.K., Bank, M.S., Siming You, S., Ok, Y., S., 2021. Interactions between microplastics, pharmaceuticals and personal care products: implications for vector transport. Environ. Int. 149, 106367 https://doi.org/10.1016/j.envint.2020.106367.

Basu, P., Sarangapani, R., Sivasubramanian, K., Venkatraman, B., 2015. Estimation of annual effective dose rate due to the ingestion of the primordial radionuclide ^{40}K for the population around the Kalpakkam nuclear site, Tamil Nadu, India. Radiat. Prot. Environ. 38, 14–22. https://doi.org/10.4103/0972-0464.162827.

Byun, J.-I., Rho, J.-H., Choi, S.-W., 2021. A shipboard real-time gamma-ray measurement system for detecting radionuclides in seawater. Nucl. Instrum. Methods. Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 1005, 165374 https://doi.org/10.1016/j.nima.2021.165374.

Cox, K.D., Covernton, G.A., Davies, H.L., Dower, J.F., Juanes, F., Dudas, S.E., 2019. Human consumption of microplastics. Environ. Sci. Technol. 53 (12), 7068–7074. https://doi.org/10.1021/acs.est.9b01517.

Darmoul, B., 1988. Pollution dans le Golfe de Gabès (Tunisie): bilan des six années de surveillance (1976-1981). Bull. Inst. Natl. Sci. Technol. Mer. Salam 15, 61–85.

Darmoul, B., Hadj Ali Salem, M., Vitiello, P., 1980. Effets des rejets industriels de la région de Gabès (Tunisie) sur le milieu récepteur. Bull. Inst. Natl. Sci. Technol. Mer. Salam 7, 5–61.

Delacroix, D., Guerre J.P., Leblanc, P., 2006 Guide Pratique Radionucleide & Radioprotection, Deuxième éd., CEA-EDP Science.

EIB, European Investment Bank, 2004. ⟨https://www.eib.org/en/press/all/2004–076-eur-34-mio-for-environmental-improvements-in-the-coastal-area-of-tunisia⟩. (Accessed 13 May 2022).

El Kateb, A., Stalder, C., Neururer, C., Pisapia, C., Spezzaferri, S., 2016. Correlation between pollution and decline of Scleractinian *Cladocora caespitosa* (Linnaeus, 1758) in the Gulf of Gabes. Heliyon 2, e00195. https://doi.org/10.1016/j.heliyon.2016.e00195.

El Kateb, A., Stalder, C., Rüggeberg, A., Neururer, C., Spangenberg, J.E., Spezzaferri, S., 2018. Impact of industrial phosphate waste discharge on the marine environment in the Gulf of Gabes (Tunisia. PLoS One 13 (5), e0197731. https://doi.org/10.1371/journal.pone.0197731.

El Zrelli, R. , 2017. Metallic Trace Element Transfer Modalities in the Central Part of Gabes Gulf, Tunisia: a Geochemical, Mineralogical, Sedimentological and Biological Approach (PhD dissertation), University of Toulouse III-Paul Sabatier, France.

El Zrelli, R., Baliteau, J.Y., Yacoubi, L., Castet, S., Grégoire, M., Fabre, S., Sarazin, V., Daconceicao, L., Courjault-Radé, P., Rabaoui, L., 2021. Rare earth elements characterization associated to the phosphate fertilizer plants of Gabes (Tunisia, Central Mediterranean Sea): geochemical properties and behavior, related economic losses, and potential hazards. Sci. Total Environ. 791C, 148268 https://doi.org/10.1016/j.scitotenv.2021.148268.

El Zrelli, R., Courjault-Radé, P., Rabaoui, L., Castet, S., Michel, S., Bejaoui, N., 2015. Heavy metal contamination and ecological risk assessment in the surface sediments of the coastal area surrounding the industrial complex of Gabes city Gulf of Gabes, SE Tunisia. Mar. Pollut. Bull. 101, 922–929. https://doi.org/10.1016/j.marpolbul.2015.10.047.

El Zrelli, R., Courjault-Radé, P., Rabaoui, L., Daghbouj, N., Mansour, L., Balti, R., Castet, S., Attia, F., Michel, S., Bejaoui, N., 2017. Biomonitoring of coastal pollution in the Gulf of Gabes (SE. Tunisia): use of *Posidonia oceanica* seagrass as a bioindicator and its mat as an archive of coastal metallic contamination. Environ. Sci. Pollut. Res. 24, 22214–22225. https://doi.org/10.1007/s11356-017-9856-x.

El Zrelli, R., Rabaoui, L., Ben Alaya, M., Daghbouj, N., Castet, S., Besson, P., Michel, S., Bejaoui, N., Courjault-Radé, P., 2018b. Seawater quality assessment and identification of pollution sources along the central coastal area of Gabes Gulf (SE Tunisia): Evidence of industrial impact and implications for marine environment protection. Mar. Pollut. Bull. 127, 445–452. https://doi.org/10.1016/j.marpolbul.2017.12.012.

El Zrelli, R., Rabaoui, L., Daghbouj, N., Abda, H., Castet, S., Josse, C., van Beek, P., Souhaut, M., Michel, S., Bejaoui, N., Courjault-Radé, P., 2018a. Characterization of phosphate rock and phosphogypsum from Gabes phosphate fertilizer factories (SE Tunisia): high mining potential and implications for environmental protection. Environ. Sci. Pollut. Res. 25, 14690–14702. https://doi.org/10.1007/s11356-018-1648-4.

El Zrelli, R., Rabaoui, L., Abda, H., Daghbouj, N., Pérez-López, R., Castet, S., Aigouy, T., Bejaoui, N., Courjault-Radé, P., 2019b. Characterization of the role of phosphogypsum foam in the transport of metals and radionuclides in the Southern Mediterranean Sea. J. Hazard. Mater. 363, 258–267. https://doi.org/10.1016/j.jhazmat.2018.09.083.

El Zrelli, R., Rabaoui, L., Ben Alaya, M., Castet, S., Zouiten, C., Bejaoui, N., Courjault-Radé, P., 2019c. Decadal effects of solid industrial wastes on the coast: Gulf of Gabes (Tunisia, Southern Mediterranean Sea) as an example. In: Estuar. Coast. Shelf Sci., 224, pp. 281–288. https://doi.org/10.1016/j.ecss.2019.04.021.

El Zrelli, R., Rabaoui, L., Roa-Ureta, R.H., Gallai, N., Castet, S., Grégoire, M., Bejaoui, N., Courjault-Radé, P., 2020. Economic impact of human-induced shrinkage of *Posidonia oceanica* meadows on coastal fisheries in the Gabes Gulf (Tunisia, Southern Mediterranean Sea). Mar. Pollut. Bull. 55, 111124 https://doi.org/10.1016/j.marpolbul.2020.111124.

El Zrelli, R., Rabaoui, L., Van Beek, P., Castet, S., Souhaut, M., Grégoire, M., Courjault-Radé, P., 2019a. Natural radioactivity and radiation hazard assessment of industrial wastes from the coastal phosphate treatment plants of Gabes (Tunisia, Southern Mediterranean Sea. Mar. Pollut. Bull. 146, 454–461. https://doi.org/10.1016/j.marpolbul.2019.06.075.

Fotopoulou, K.N., Karapanagioti, H.K., 2015. Surface properties of beached plastics. Environ. Sci. Pollut. Res. 22, 11022–11032. https://doi.org/10.1016/j.marenvres.2012.08.010.

Gao, X., Hassan, I., Peng, Y., Huo, S., Ling, L., 2021. Behaviors and influencing factors of the heavy metals adsorption onto microplastics: a review. J. Clean. Prod. 319, 128777 https://doi.org/10.1016/j.jclepro.2021.128777.

Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. Sci. Adv. 3 (7), e1700782 https://doi.org/10.1126/sciadv.1700782.

Guerrero, J.L., Gutiérrez-Álvarez, I., Hierro, A., Pérez-Moreno, S.M., Olías, M., Bolívar, J. P., 2021. Seasonal evolution of natural radionuclides in two rivers affected by acid mine drainage and phosphogypsum pollution. Catena 197, 104978. https://doi.org/10.1016/j.catena.2020.104978.

Haase, G., Vagt, T., Fritsche, J., 2021. Monitoring environmental radionuclide activity of the incident-relevant radionuclides ^{137}Cs und ^{90}Sr in animal feed and foodstuffs consumed in Germany. J. Consum. Prot. Food Saf. 16, 97–101. https://doi.org/10.1007/s00003-020-01309-7.

Hamza, F., Kahli, A., Chokri, M.-A., Almalki, M., Hanane, S., 2021. Urban and industrial landscapes interact with microhabitat to predict occurrence of European Turtle Dove (*Streptopelia turtur*) in Mediterranean oases: implications for conservation. Landsc. Urban Plan. 215, 104219 https://doi.org/10.1016/j.landurbplan.2021.104219.

Heldal, H.E., Helvik, L., Haanes, H., Volynkin, A., Jensen, H., Lepland, A., 2021. Distribution of natural and anthropogenic radionuclides in sediments from the Vefsnfjord, Norway. Mar. Pollut. Bull. 172, 112822 https://doi.org/10.1016/j.marpolbul.2021.112822.

Huang, J., Chen, H., Zheng, Y., Yang, Y., Zhang, Y., Gao, B., 2021. Microplastic pollution in soils and groundwater: characteristics, analytical methods and impacts. Chem. Eng. J. 425, 131870 https://doi.org/10.1016/j.cej.2021.131870.

ICRP, The International Commission on Radiological Protection, 2008. Radiation Dose to Patients From Radiopharmaceuticals. Addendum 3 to ICRP Publication 53. ICRP Publication, 106. Ann. ICRP 38, 1–2. ⟨https://www.icrp.org/publication.asp?id=icrp%20publication%20106⟩. (Accessed 24 June 2021).

ICRP, The International Commission on Radiological Protection, 2012. Compendium of dose coefficients based on ICRP Publication 60. ICRP Publication 119. Ann. ICRP 41 (Suppl). ⟨https://www.icrp.org/publication.asp?id=ICRP%20Publication%20119⟩.

Ioannidis, I., Anastopoulos, I., Pashalidis, I., 2022. Microplastics as radionuclide (U-232) carriers. J. Mol. Liq. 351, 118641 https://doi.org/10.1016/j.molliq.2022.118641.

Issac, M.N., Kandasubramanian, B., 2021. Effect of microplastics in water and aquatic systems. Environ. Sci. Pollut. Res. 28, 19544–19562. https://doi.org/10.1007/s11356-021-13184-2.

Johansen, M.P., Prentice, E., Cresswell, T., Howell, N., 2018. Initial data on adsorption of Cs and Sr to the surfaces of microplastics with biofilm. J. Environ. Radioact. 190–191, 130–133. https://doi.org/10.1016/j.jenvrad.2018.05.001.

Joo, S.H., Liang, Y., Kim, M., Byun, J., Heechul Choi, H., 2021. Microplastics with adsorbed contaminants: mechanisms and Treatment. Environ. Chall. 3, 100042 https://doi.org/10.1016/j.envc.2021.100042.

Kang, T.-W., Han, Y.-U., Na, E.H., Koo, B.-J., Park, W.-P., 2021. Deposition of Fukushima nuclear power plant accident-derived radiocesium in the soils of Jeju Island, Korea, and evidence for long- and short-lived radionuclides in rainwater. Chemosphere 264, 128457. https://doi.org/10.1016/j.chemosphere.2020.128457.

Kedzierski, M., D'Almeida, M., Magueresse, A., Le Grand, A., Duval, H., César, G., Sire, O., Bruzaud, S., Le Tilly, V., 2018. Threat of plastic ageing in marine environment. Adsorption/desorption of micropollutants. Mar. Pollut. Bull. 127, 684–694. https://doi.org/10.1016/j.marpolbul.2017.12.059.

Koelmans, A.A., Redondo-Hasselerharm, P.E., Nor, N.H.M., de Ruijter, R., Mintening, S. M., Kooi, M., 2022. Risk assessment of microplastic particles. Nat. Rev. Mater. 7, 138–152. https://doi.org/10.1038/s41578-021-00411-y.

Koutnik, V.S., Leonard, J., Alkidim, S., DePrima, F.J., Ravi, S., Hoek, E.M.V., Mohanty, S. K., 2021. Distribution of microplastics in soil and freshwater environments: global analysis and framework for transport modeling. Environ. Pollut. 274, 116552 https://doi.org/10.1016/j.envpol.2021.116552.

Li, H., Wang, F., Li, J., Deng, S., Zhang, S., 2021. Adsorption of three pesticides on polyethylene microplastics in aqueous solutions: kinetics, isotherms, thermodynamics, and molecular dynamics simulation. Chemosphere 264, 128556. https://doi.org/10.1016/j.chemosphere.2020.128556.

Lionetto, F., Corcione, C.E., 2021. An overview of the sorption studies of contaminants on poly(ethylene terephthalate) microplastics in the marine environment. J. Mar. Sci. Eng. 9, 445. https://doi.org/10.3390/jmse9040445.

Makon, T.B., Nemba, R.M., Tchokossa, P., 2011. Investigation of gamma-emitting natural radioactive contents in three types of Vernonia consumed in Cameroon. World J. Nucl. Sci. Technol. 1, 37–45. https://doi.org/10.4236/wjnst.2011.12007.

Mao, R., Song, J., Yan, P., Ouyang, Z., Wu, R., Liu, S., Guo, X., 2021. Horizontal and vertical distribution of microplastics in the Wuliangsuhai Lake sediment, northern China. Sci. Total Environ. 754, 142426 https://doi.org/10.1016/j.scitotenv.2020.142426.

Martin, C., Baalkhuyur, F., Valluzzi, L., Saderne, V., Cusack, C., Almahasheer, H., Krishnakumar, P.K., Rabaoui, L., Qurban, M.A., Arias Ortiz, A., Masqué, P., Duarte, C.M., 2020. Exponential increase of plastic burial in mangrove sediments as a major plastic sink. Sci. Adv. 6 (44), eaaz5593.

Moog, D., Schmitt, J., Senger, J., Zarzycki, J., Rexer, K.H., Linne, U., Erb, T., Maier, U.G., 2019. Using a marine microalga as a chassis for polyethylene terephthalate (PET) degradation. Microb. Cell Factor. 18 (1), 171. https://doi.org/10.1186/s12934-019-1220-z.

Niu, L., Li, Y., Li, Y., Hu, Q., Wang, C., Hu, J., Zhang, W., Wang, L., Zhang, C., Huanjun Zhang, H., 2021. New insights into the vertical distribution and microbial degradation of microplastics in urban river sediments. Water Res. 188, 116449 https://doi.org/10.1016/j.watres.2020.116449.

Nor, N.H.M., Kooi, M., Diepens, N.J., Koelmans, A.A., 2021. Lifetime accumulation of microplastic in children and adults. Environ. Sci. Technol. 55 (8), 5084–5096. https://doi.org/10.1021/acs.est.0c07384.

Pergent, G., Kempf, M., 1993 L'environnement Marin Côtier en Tunisie, IFREMER, Brest, France. ⟨https://archimer.ifremer.fr/doc/00105/21592/⟩. (Accessed 2 June 2022).

Pham, D.N., Clark, L., Li, M., 2021. Microplastics as hubs enriching antibiotic-resistant bacteria and pathogens in municipal activated sludge. J. Hazard. Mater. Lett. 2, 100014 https://doi.org/10.1016/j.hazl.2021.100014.

Puckowski, A., Cwięk, W., Mioduszewska, K., Stepnowski, P., Białk-Bielińska, A., 2021. Sorption of pharmaceuticals on the surface of microplastics. Chemosphere 263, 127976. https://doi.org/10.1016/j.chemosphere.2020.127976.

Qureshi, A.A., Tariq, S., Din, K.U., Manzoor, S., Calligaris, C., Waheed, A., 2014. Evaluation of excessive lifetime cancer risk due to natural radioactivity in the rivers sediments of Northern Pakistan. J. Radiat. Res. Appl. Sci. 7, 438–447. https://doi.org/10.1016/j.jrras.2014.07.008.

Rabaoui, L., Balti, R., El Zrelli, R., Tlig-Zouari, S., 2014. Assessment of heavy metals pollution in the Gulf of Gabes (Tunisia) using four mollusk species. Mediterr. Mar. Sci. 15, 45–58. https://doi.org/10.12681/mms.504.

Rabaoui, L., El Zrelli, R., Balti, R., Mansour, L., Courjault-Radé, P., Daghbouj, N., Tlig-Zouari, S., 2017. Metal bioaccumulation in two edible cephalopods in the Gulf of Gabes, South-eastern Tunisia: environmental and human health risk assessment. Environ. Sci. Pollut. Res. 24, 1686–1699. https://doi.org/10.1007/s11356-016-7945-x.

Rabaoui, L., El Zrelli, R., Ben Mansour, M., Balti, R., Mansour, L., Tlig-Zouari, S., Guerfel, M., 2015. On the relationship between the diversity and structure of benthic macroinvertebrate communities and sediment enrichment with heavy metals in

Gabes Gulf Tunisia. J. Mar. Biol. Assoc. UK 95, 233–245. https://doi.org/10.1017/S0025315414001489.

Ragusa, A., Svelato, A., Santacroce, C., Catalano, P., Notarstefano, V., Carnevali, O., Papa, F., Rongioletti, M.C.A., Baiocco, F., Draghi, S., D'Amore, E., Rinaldo, D., Matta, M., Giorgini, E., 2021. Plasticenta: first evidence of microplastics in human placenta. Environ. Int. 146, 106274 https://doi.org/10.1016/j.envint.2020.106274.

Rochman, C.M., Hentschel, B.T., Teh, S.J., 2014. Long-term sorption of metals is similar among plastic types: implications for plastic debris in aquatic environments. PLoS One 9 (1), e85433. https://doi.org/10.1371/journal.pone.0085433.

Sarkhel, R., Sengupta, S., Das, P., Bhowal, A., 2020. Comparative biodegradation study of polymer from plastic bottle waste using novel isolated bacteria and fungi from marine source. J. Polym. Res. 27, 16. https://doi.org/10.1007/s10965-019-1973-4.

Senathirajah, K., Attwood, S., Bhagwat, G., Carbery, M., Wilson, S., Palanisami, T., 2021. Estimation of the mass of microplastics ingested – a pivotal first step towards human health risk assessment. J. Hazard. Mater. 404 (B), 124004 https://doi.org/10.1016/j.jhazmat.2020.124004.

Sherif, M.I., Sturchio, N.C., 2021. Elevated radium levels in Nubian Aquifer groundwater of Northeastern Africa. Sci. Rep. 11, 78. https://doi.org/10.1038/s41598-020-80160-0.

Smith, M., Love, D.C., Rochman, C.M., Neff, R.A., 2018. Microplastics in seafood and the implications for human health. Curr. Environ. Health Rep. 5 (3), 375–386 https://doi.org/10.1007%2Fs40572-018-0206-z.

Song, X., Wu, X., Song, X., Shi, C., Zhang, Z., 2021. Sorption and desorption of petroleum hydrocarbons on biodegradable and nondegradable microplastics. Chemosphere 273, 128553. https://doi.org/10.1016/j.chemosphere.2020.128553.

UNSCEAR, The United Nations Scientific Committee on the Effects of Atomic Radiation, 2000. Sources and Effects of Ionizing Radiation. Report to the General Assembly with Scientific Annexes, United Nations, New York. ⟨https://www.unscear.org/docs/publications/2000/UNSCEAR_2000_Report_Vol.I.pdf⟩.

UNSCEAR, The United Nations Scientific Committee on the Effects of Atomic Radiation, 2008. Sources and Effects of Ionizing Radiation. United Nations Publication, New York. ⟨https://www.unscear.org/docs/reports/2008/11-80076_Report_2008_Annex_D.pdf⟩.

Uwatse, O.B., Olatunji, M.A., Khandaker, M.U., Amin, Y.M., Bradley, D.A., Alkhorayef, M., Alzimami, K., 2015. Measurement of natural and artificial radioactivity in infant powdered milk and estimation of the corresponding annual effective dose. Environ. Eng. Sci. 32 (10), 838–846. https://doi.org/10.1089/ees.2015.0114.

van Beek, P., Souhaut, M., Lansard, B., Bourquin, M., Reyss, J.L., von Ballmoos, P., Jean, P., 2013. LAFARA: a new underground laboratory in the French Pyrénées for ultralow-level gamma-ray spectrometry. J. Environ. Radioact. 116, 152–158. https://doi.org/10.1016/j.jenvrad.2012.10.002.

Wakkaf, T., El Zrelli, R., Kedzierski, M., Balti, R., Shaiek, M., Mansour, L., Tlig-Zouari, S., Bruzaud, S., Rabaoui, L., 2020a. Characterization of microplastics in the surface waters of an urban lagoon (Bizerte lagoon, Southern Mediterranean Sea): Composition, density, distribution, and influence of environmental factors. Mar. Pollut. Bull. 160, 111625 https://doi.org/10.1016/j.marpolbul.2020.111625.

Wakkaf, T., El Zrelli, R., Kedzierski, M., Balti, R., Shaiek, M., Mansour, L., Tlig-Zouari, S., Bruzaud, S., Rabaoui, L., 2020b. Microplastics in edible mussels from a southern Mediterranean lagoon: preliminary results on seawater-mussel transfer and implications for environmental protection and seafood safety. Mar. Pollut. Bull. 158, 111355 https://doi.org/10.1016/j.marpolbul.2020.111355.

Wakkaf, T., El Zrelli, R., Yacoubi, L., Kedzierski, M., Lin, Y.-J., Mansour, L., Bruzaud, S., Rabaoui, L., 2022. Seasonal patterns of microplastics in surface sediments of a Mediterranean lagoon heavily impacted by human activities (Bizerte lagoon, Northern Tunisia. Environ. Sci. Pollut. Res. https://doi.org/10.1007/s11356-022-21129-6.

Wang, F., Lai, Z., Peng, G., Luo, L., Liu, K., Huang, X., Xu, Y., Shen, Q., Li, D., 2021. Microplastic abundance and distribution in a Central Asian desert. Sci. Total Environ. 800, 149529 https://doi.org/10.1016/j.scitotenv.2021.149529.

WB, Word Bank, 2022. Life Expectancy at Birth. ⟨https://data.worldbank.org/indicator/SP.DYN.LE00.IN?locations=TN⟩. (Accessed 17 June 2022).

Yang, W.L., Neoh, K.G., Kang, E., Teo, S.M., Rittschof, D., 2014. Polymer brush coatings for combating marine biofouling. Prog. Polym. Sci. 39, 1017–1042. https://doi.org/10.1016/j.progpolymsci.2014.02.002.

Zayen, A., Sayadi, S., Chevalier, C., Boukthir, M., Ben Ismail, S., Tedetti, M., 2020. Microplastics in surface waters of the Gulf of Gabes, southern Mediterranean Sea: distribution, composition and influence of hydrodynamics. Estuar. Coast. Shelf Sci. 242, 106832 https://doi.org/10.1016/j.ecss.2020.106832.

Zhang, N., Li, Y.B., He, H.R., Zhang, J.F., Ma, G.S., 2021. You are what you eat: Microplastics in the feces of young men living in Beijing. Sci. Total Environ. 767, 144345 https://doi.org/10.1016/j.scitotenv.2020.144345.