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Radiological risks from 40K, 226Ra and 232Th in urbanised and industrialised karstic coastal area (Kaštela Bay, Croatia)

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Abstract:	Radiological risks associated with 40 K, 226 Ra and 232 Th massic activities in limestones, marls, stream sediments, and soils of the Kaštela Bay (Croatia) coastal area were assessed by calculating outdoor absorbed dose rates in air (D), annual outdoor effective dose rates (D ef), radium equivalent activities (Ra eq), and external hazard indices (H ex). Radionuclides relative contributions to D and H ex were determined for all four types of samples as well as their total contribution to H ex in all samples. D , D ef , Ra eq , and H ex were the lowest in limestones and the highest in soils. Maximum Ra eq and H ex in soil were below the recommended values of 370 Bq/kg and 1.0. No adverse radiological effects were determined in the researched area. The most important contribution to D and H ex in limestones was almost exclusively from 226 Ra, in marls from 40 K, in stream sediments from 226 Ra and in soils from 232 Th. The most significant total contribution to H ex in all samples came from 226 Ra and 232 Th, and the lowest came from 40 K. 226 Ra showed the largest variability of its total contribution to H ex , with tendency to higher values. Special attention should be given to 226 Ra when studying radiological risks in typical karstic areas, irrespectively of other possible influences of geological background.
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- You need to clearly indicate that statistic has been used in each case and report adequately the p value. For p values lower than 0.000001 must be reported as $p < 0.0001$. The p value cannot be equal to 0 (zero), when a statistical software reports a p value of 0, indicates that the value is very low and must be reported as < 0.0001.

Response: *p*-values are clearly indicated wherever they were determined and they are corrected where necessary. *p*-values are added in *Data analysis* chapter regarding Shapiro-Wilk's W test. Values equal to zero are corrected into $p < 0.0001$ in the *Radiological risks* chapter regarding ANOVA results. *p*-value is added in the *Discussion* chapter when mentioning the lack of statistical difference between ^{226}Ra and ^{232}Th contributions.

Zagreb, 18th February 2022

Dear Editor,

after performed revisions, I am resubmitting the manuscript titled: **Radiological risks from ^{40}K, ^{226}Ra and ^{232}Th in urbanised and industrialised karstic coastal area (Kaštela Bay, Croatia)** by I. Lovrenčić Mikelić and D. Barišić.

All reviewer's comments have been addressed. Actions taken considering reviewer's comment are listed in response to reviewers.

I hope that the revised manuscript will now be appropriate for publication in the Environmental Science and Pollution Research journal.

Best regards.

dr. sc. Ivanka Lovrenčić Mikelić

Radiological risks from ^{40}K, ^{226}Ra and ^{232}Th in urbanised and industrialised karstic coastal area (Kaštela Bay, Croatia)

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Abstract

Radiological risks associated with ^{40}K, ^{226}Ra and ^{232}Th massic activities in limestones, marls, stream sediments, and soils of the Kaštela Bay (Croatia) coastal area were assessed by calculating outdoor absorbed dose rates in air (D), annual outdoor effective dose rates (D_{ef}), radium equivalent activities ($\mathrm{Ra_{eq}}$), and external hazard indices (H_{ex}). Radionuclides relative contributions to D and H_{ex} were determined for all four types of samples as well as their total contribution to H_{ex} in all samples. D, D_{ef}, $\mathrm{Ra_{eq}}$, and H_{ex} were the lowest in limestones and the highest in soils. Maximum $\mathrm{Ra_{eq}}$ and H_{ex} in soil were below the recommended values of 370 Bq/kg and 1.0. No adverse radiological effects were determined in the researched area. The most important contribution to D and H_{ex} in limestones was almost exclusively from ^{226}Ra, in marls from ^{40}K, in stream sediments from ^{226}Ra and in soils from ^{232}Th. The most significant total contribution to H_{ex} in all samples came from ^{226}Ra and ^{232}Th, and the lowest came from ^{40}K. ^{226}Ra showed the largest variability of its total contribution to H_{ex}, with tendency to higher values. Special attention should be given to ^{226}Ra when studying radiological risks in typical karstic areas, irrespectively of other possible influences of geological background.

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Keywords: Adriatic Sea; karst; limestones; marl; radiological risk; soil; stream sediment

Introduction

According to data for 2019, population around the Kaštela Bay was approximately 250 000 (Croatian Bureau of Statistics 2019). Towns around the Kaštela Bay are merged into one urban agglomeration, the biggest one on the east Adriatic Sea coast. It includes the City of Split that is the second most populated city in Croatia and the first one in Dalmatia region. Numerous industrial and agricultural activities were and are still present around the Kaštela Bay (Lovrenčić Mikelić et al. 2021) but the most important one from the radioactivity point of view is the former "Jugovinil"/"Adriavinil" chemical factory that operated in the 1950 – 1990 period. The factory used coal with elevated natural radioactivity in its thermoelectric power plant, which resulted in production of bottom and fly ash with elevated ^{238}U, ^{235}U and ^{226}Ra massic activities and which was characterised as TENORM (technologically enhanced naturally occurring radioactive material) (Lovrencic et al. 2007; Marović and Senčar 1999; Marović et al. 2006; Orescanin et al. 2005). This TENORM was deposited both in regulated and in unregulated manner near the factory and partly even in the sea. TENORM from other locations was also deposited here. It was estimated that 50 000 t of bottom and fly ash were deposited on the area of 18 000 m^2 (Marović et al. 2006).

The TENORM deposition site might be considered a possible source of radionuclides for the Kaštela Bay environment, both terrestrial and marine. This is especially important because the deposition site is located in a karstic coastal area and because these kinds of environments are very sensitive to all contaminants/pollutants. Furthermore, this is even more important when such areas are densely populated. Numerous studies related to radioactivity around the Kaštela Bay were performed (Lovrencic et al. 2005, 2007; Lovrenčić Mikelić et al. 2021; Marović and Senčar 1999; Orescanin et al. 2006; Skoko et al. 2014, 2017, 2019).

However, only few assessed radiological risks (Marović and Senčar 1999; Skoko et al. 2019) and these risks were studied only for the TENORM deposition site. Other environmentally or geologically relevant matrices (different rocks and sediments) were not studied. The same partial, or specifically targeted, approach is regularly applied in studies performed in other researched areas. Furthermore, studies encompassing limestones, marls and related soils and/or stream sediments as a unique system in which "contrasting parent materials" exist are rare (Gaspar et al. 2021; Lovrenčić Mikelić et al. 2021) and they did not address radiological risks.

Therefore, a karstic coastal area potentially influenced by anthropogenic activity is studied as a whole in the presented study and all geologically relevant rocks and sediments were considered. These were limestones, marls, stream sediments and soils. Studied in such a way, the researched area may serve as a model area for other similar karstic environments. Since ^{40}K, ^{232}Th, ^{238}U and their decay products are the primary source of natural background radiation and human exposure from terrestrial sources, ^{40}K, ^{226}Ra and ^{232}Th related radiological risks were studied. The study presented in this paper was conducted to obtain deeper insight into radiological risks related to individual types of samples and radionuclides. Radiological risks are considered here from geological perspective alongside health perspective. Results of this study can be useful for future researches of karstic terrestrial environments and for local decision making regarding radiological protection of the population.

Material and methods

Study area

Kaštela Bay is a semi-enclosed bay on the eastern coast of the Adriatic Sea, Croatia (Fig. 1). It is enclosed by the narrow coastal plain with mountains in the hinterland to the north, by the Čiovo Island to the southwest and by the City of Split to the southeast (Fig. 1). The area from the Trogir town to the west, through the Kaštela town, and to the City of Split to the east is merged into one urban agglomeration with pronounced industrialisation. Čiovo Island is much less urbanized and without industrial activities. It is connected with mainland by a bridge.

The researched area is part of the central Adriatic Sea area that is characterised with a Csa climate type according to Köppen climate classification (Gajić-Čapka and Zaninović 2008). Mean annual air temperature is 17 °C, with its maximum in summer (mostly in July and less often in August) and minimum in January (Zaninović 2008). Mean annual precipitation amount is in 800 – 900 mm/year range. The lowest precipitation amount occurs in warm part of the year (April to September) with the main minimum in July (Gajić-Čapka et al. 2008). The driest month receives less than 40 mm of precipitation. Dry spells longer than 10 days occur and are most frequent from June to September. The main precipitation maximum is in November.

Jadro River is the only permanent surface stream flowing into the Bay near the Solin town. Many temporary streams occur on the north coastal plain, but they depend on the amount of precipitation (Lovrenčić Mikelić et al. 2013). Due to irregular and very often scarce precipitation, these streams dry up frequently. Nor permanent neither temporary streams in form of rivers or brooks exist on the Čiovo Island.

Simplified lithological composition of the Kaštela Bay coastal area is presented in Fig. 1. Carbonate rocks and varieties of Eocene flysch and flysch-like rocks are two most important groups of rocks building up the area (Magaš and Marinčić 1973; Marinčić et al. 1971). Flysch and flysch-like rocks are the most abundant rocks along the north coast of the

Bay and on the Split peninsula. Lenses of marl and limestone are present in these rocks. Carbonate rocks predominate on the Čiovo Island and they are of some importance on the Marjan hill and around Divulje. Carbonate rocks include Cretaceous limestones and limestone dolomites (Čiovo Island), Eocene foraminiferal limestones (Marjan hill), and Pleistocene breccias consisting of fragments of carbonate sediment bound by bauxitic matrix (around Divulje) (Magaš and Marinčić 1973; Marinčić et al. 1971).

Sampling

Forty-five samples of rocks and sediments of the Kaštela Bay coastal area were collected in 2005 – 2008 period. Limestones s.l. (K), marls (L), stream sediments (PS), and soils (T) were sampled. Limestones s.l. refer to all rocks of limestone composition irrespectively of their genesis. Soils are regarded as residual clastic sediments (Tišljar 1994). Sampling locations are presented in Fig. 1 and sampling locations coordinates are given in Table 1.

One to two kg of samples were collected at each sampling location. Samples were placed into labelled plastic bags and transported to laboratory. Limestones s.l. and marls were sampled on accessible open outcrops. Stream sediments were sampled in brook beds (0 – 10 cm depth) on the north coast of the Kaštela Bay. Due to non-existent brooks or rivers on the Čiovo Island, surface samples (0 – 10 cm depth) of undisturbed soils were collected there. Soil sampling locations where chosen in relation to soil transport to the sea by precipitation water. Such soil transport is equivalent to stream sediment transport by brooks or rivers. Soil samples were taken where soil transport by precipitation was observed. Sample T2 was brown soil and all other soil samples were *terra rossa*. More details on sampling are given in Lovrenčić Mikelić et al. (2013, 2021).

Gamma-spectrometry

Stream sediment and soil samples were first dried at 105 °C overnight to achieve constant mass, then ground in a mill with agate spherules or in an agate mortar, and homogenized. Limestone s.l. and marl samples were first crushed in a crusher, then dried overnight at 105 °C, and homogenized. Afterwards, all samples were stored in plastic containers of 125 cm^3 volume, closed with lids and weighted. The containers lids were sealed with a self-adhesive tape and stored for at least four weeks to allow ingrowth of gaseous ^{222}Rn.

Gamma-spectrometric analysis was preformed using HPGe coaxial (relative efficiency: 25.3 %, resolution at 1332.5 keV (^{60}Co): 1.75 keV) and InSpector 2000 (relative efficiency: 25.4 %, resolution at 1332.5 keV (^{60}Co): 1.80 keV) detectors. Detectors were coupled with multichannel analysers with 8192 channels (Canberra Industries). Spectra were collected for 80 000 s and analysed with Genie 2000 software package (Canberra Industries 2006). Reference date for recalculation of all massic activities was 6th June 2005. ^{40}K massic activity was directly determined from its 1460.75 keV peak. ^{226}Ra weighted mean activity was calculated using its progenies (^{214}Pb: 295.21 keV and 351.92 keV peaks; ^{214}Bi: 609.31 keV, 1120.28 keV and 1764.49 keV peaks) and assuming secular equilibrium between ^{226}Ra and its progenies (Canberra Industries 2006; Saïdou et al. 2008). ^{232}Th weighted mean activity was calculated using ^{212}Pb (238.63 keV peak) and ^{228}Ac (338.32 keV, 911.20 keV and 968.97 keV peaks). Information about energy and efficiency calibrations and quality control can be found in Lovrenčić Mikelić et al. (2017).

Calculation of radiological risks

Outdoor absorbed dose rate in air *D* (in nGy/h) was calculated according to expression (Örgün et al. 2007; UNSCEAR 2000, 2008):

$D = \sum_{x} C_x \times a_x$, i.e. (1)

$D = 0.462 \times a(^{226}\mathrm{Ra}) + 0.604 \times a(^{232}\mathrm{Th}) + 0.0417 \times a(^{40}\mathrm{K})$ (2)

where: C_x – dose conversion factor for respective radionuclide (nGy/h for Bq/kg), a_x and $a(x)$ – massic activity of respective radionuclide (Bq/kg).

Annual outdoor effective dose rate D_{ef} (in mSv/y) was calculated according to the following formula (Örgün et al. 2007; UNSCEAR 2000; Yang et al. 2005):

$D_{ef} = D \times t \times O_f \times C_f \times 10^{-6}$ (3)

where: D – outdoor absorbed dose rate in air (nGy/h), t – annual number of hours approximated as 8760 (h), O_f – outdoor occupancy factor (under assumption that 20 % of the total annual time is spent outdoors, i.e. $O_f = 0.2$), C_f – conversion factor ($C_f = 0.7$ Sv/Gy).

Radium equivalent activity Ra_{eq} (in Bq/kg) was calculated as (Farai and Ademola 2005; Mavi and Akkurt 2010; Ngachin et al. 2007):

$Ra_{eq} = a(^{226}\mathrm{Ra}) + 1.43 \times a(^{232}\mathrm{Th}) + 0.077 \times a(^{40}\mathrm{K})$ (4)

where: $a(^{226}\mathrm{Ra})$, $a(^{232}\mathrm{Th})$, $a(^{40}\mathrm{K})$ – ^{226}Ra, ^{232}Th and ^{40}K massic activities (Bq/kg), respectively. Ra_{eq} should be lower than the recommended value of 370 Bq/kg (Mavi and Akkurt 2010; Ngachin et al. 2007).

External hazard index H_{ex} was calculated as follows (El-Arabi 2007; Farai and Ademola 2005; Mavi and Akkurt 2010):

$H_{ex} = a(^{226}\mathrm{Ra})/370 + a(^{232}\mathrm{Th})/259 + a(^{40}\mathrm{K})/4810$ (5)

where: $a(^{226}\mathrm{Ra})$, $a(^{232}\mathrm{Th})$, $a(^{40}\mathrm{K})$ – ^{226}Ra, ^{232}Th and ^{40}K massic activities (Bq/kg), respectively. H_{ex} should be less or equal to unity.

Data analysis

Results were summarised by means of descriptive statistics. Range, mean value, median and standard deviation were given for radionuclides massic activities and radiological parameters. Radionuclides total contribution to external hazard index was presented by Box and Whisker plots using Statistica 7.0 software (StatSoft, Inc. 2004). Additionally, Shapiro-Wilk's W test (significance level: $p < 0.05$) was performed to test data distribution ("normality of data") by the same software. Since the data for all tested radionuclides were not normally distributed (^{226}Ra: $p < 0.0001$, ^{232}Th: $p = 0.00002$, ^{40}K: $p = 0.04342$) and that their distributions were skewed even after log-transformation (^{226}Ra: $p = 0.00011$, ^{232}Th: $p < 0.0001$, ^{40}K: $p < 0.0001$), one-way non-parametric ANOVA (analysis of variance) or Kruskal-Wallis test, followed by a post-hoc test, was performed using Statistica 7.0 (StatSoft, Inc. 2004). Post-hoc test compared mean ranks of all pairs of groups giving *p*-values (two-sided significance levels) as a result (StatSoft, Inc. 2004). Statistically significant difference was defined as $p < 0.05$. It should be noted that inferential statistics results should be taken as preliminary ones due to relatively small number of samples.

Results

Radionuclides massic activities

Basic statistical parameters of ^{40}K, ^{226}Ra and ^{232}Th massic activities in limestones s.l., marls, stream sediments, and soils of the Kaštela Bay costal area are given in Table 2. The lowest radionuclides activities were found mainly in limestones s.l. or in marls, and the highest in soils. The highest mean values and medians in individual sample types were observed for ^{226}Ra activities in limestones s.l. and for ^{40}K activities in all other sample types.

Radiological risks

Results of descriptive statistics for outdoor absorbed dose rate in air (D), annual outdoor effective dose rate (D_{ef}), radium equivalent activity ($\mathrm{Ra_{eq}}$) and external hazard index (H_{ex}) are presented in Table 3. All calculated parameters were the lowest in limestones s.l. and the highest in soils and ascended in the following order in different types of samples: K, L, PS and T. Medians for all risk parameters for soil were 3.7 – 3.9 times higher than for stream sediments and even approx. 12 times higher than for limestones s.l.

Relative contributions of ^{40}K, ^{232}Th and ^{226}Ra to outdoor absorbed dose rates in air and to external hazard indices in all four sample types is shown in Fig. 2. A striking difference is observed between limestones s.l. and other sample types. ^{226}Ra alone contributes almost 90 % to both D and H_{ex} in limestones s.l., while in other samples ranges for ^{226}Ra contribution are 29 % – 39 % and 30 % – 39 %, respectively. ^{40}K and ^{232}Th contributions are almost negligible to both D and H_{ex} in limestones s.l., with all values being less than 10 %. Contribution of all three radionuclides to D and H_{ex} in other sample types is more balanced, with maximum summary ranges of 21 % – 40 % and 18 % – 43 %, respectively. ^{40}K is the main contributing radionuclide to D and H_{ex} in marls (40 % and 36 %, respectively). Its contribution is more pronounced in D than in H_{ex}, when compared to ^{232}Th and ^{226}Ra contributions in marls. Stream sediments and soils show some similarities. The lowest observed contributions are from ^{40}K in both D and H_{ex} (21 % – 28 % and 18 % – 24 %, respectively). ^{226}Ra and ^{232}Th contributions are very similar for D and H_{ex} in both stream sediments and soils. Their individual contributions to D are 34 % – 40 % and 37 % – 43 % to H_{ex}. Decreasing contribution of ^{40}K to both D and H_{ex} is observed in the following order of samples: L, PS, T. Equally, contribution of ^{226}Ra and ^{232}Th increased in the same order. ^{226}Ra and ^{232}Th contributions to D and H_{ex} in soils are almost equal, with somewhat higher ^{232}Th contributions. Soil is the only sample type where ^{232}Th contribution is dominant, although not markedly, especially for H_{ex}.

Individual total contribution of ^{226}Ra, ^{232}Th and ^{40}K to H_{ex} in all collected samples is presented in Fig. 3. ^{226}Ra contribution presented the highest mean value (49 %) and median

(39 %) and the largest variability. Respective values for ^{232}Th are 31 % and 36 % and for ^{40}K both are 21 %. Maximum value for ^{226}Ra is 98 %, while for ^{232}Th and ^{40}K these values are 49 % and 39 %, respectively. The lowest contribution comes from ^{40}K with the minimum value of only 0,40 %. ^{226}Ra contribution presented tendency towards higher values, while ^{232}Th contribution presented tendency towards lower values. ANOVA showed statistically significant difference ($p < 0.0001$) in radionuclides contributions. Post-hoc test showed that ^{40}K contribution was statistically significantly different from ^{226}Ra and ^{232}Th contributions (^{40}K–^{226}Ra: $p < 0.0001$, ^{40}K–^{232}Th: $p = 0.000137$), while there was no statistical difference between ^{226}Ra and ^{232}Th ($p = 0.067374$).

Discussion

Only D and D_{ef} from the Kaštela Bay soils (Table 3) exceeded the average values given by UNSCEAR (2000) (58 nGy/h and 0.07 mSv/y, respectively). Almost all soil samples exceeded these values. Increased D and D_{ef} are attributed to natural local variability of the soil composition and to the carbonate bedrock on which the researched soils were developed. Naturally moderately increased ^{226}Ra and ^{232}Th activities of the Kaštela Bay soils were previously documented by Skoko et al. (2014). All radium equivalent activities were lower than the recommended value of 370 Bq/kg (Mavi and Akkurt 2010; Ngachin et al. 2007) and all external hazard indices were lower than unity, although maximum H_{ex} in soils was close to it (0.93) (Table 3).

Radiological risks that are the most pronounced in soils are especially associated with increased ^{226}Ra and ^{232}Th activities in soils (Tables 2 and 3). This is also supported by significantly higher contributions of ^{226}Ra and ^{232}Th to D and H_{ex} than the one of ^{40}K (Fig. 2). Taking into account that *terra rossa* soil is a residual soil developed on carbonate bedrock and that ^{226}Ra is often found in carbonates (Cowart and Burnett 1994), significant ^{226}Ra contribution was expected. Although limestones usually contain little to none ^{232}Th (Gascoyne 1982; Navas et al. 2002), thorium's strong preferential sorption to soil particles facilitates its accumulation in soils, including the *terra rossa*.

Pronounced contribution of ^{226}Ra to D and H_{ex} in limestones s.l. and almost negligible contribution of ^{40}K and ^{232}Th (Fig. 2) point to almost pure carbonate rock with negligible influence of detritic particles. Maximum ^{40}K contribution (relative to other two radionuclides) to D and H_{ex} observed only in marls (Fig. 2) reflects high content of clay minerals typical for marls, but not for other studied sample types. It implies a decrease in clay minerals content from marls to soils. This is in accordance with increasing ^{232}Th and ^{226}Ra contributions to both D and H_{ex} from marls to soils (Fig. 2) that show increasing influence of detritic/terrestrial particles other than clay minerals, including increasing carbonate influence. The highest ^{232}Th contribution to both D and H_{ex} in soils implies that detritic particles will preferably accumulate in soils.

Significant importance of carbonate bearing rocks and sediments in the researched area considering radiological risks was also supported by Fig. 3, where it was shown that ^{226}Ra was one of the most important total contributors to H_{ex} in all samples considered together. ^{226}Ra contribution tendency to higher values may be ascribed to strong influence of carbonates s.l. (Fig. 2). Comparable influence of ^{232}Th bearing detritic particles is supported by the lack of statistical difference between ^{226}Ra and ^{232}Th contributions ($p = 0.067374$). However, ^{232}Th contribution tendency towards lower values implies strong influence of pure carbonate rocks. The lowest ^{40}K total contribution to H_{ex} in all samples (Fig. 3) also supports the observed lesser importance of clay bearing rocks and sediments of the Kaštela Bay coastal area for radiological risks. This may be applied to outdoor absorbed dose rates in air as well since both H_{ex} and D presented the same patterns in all four types of sample when studied individually (Fig. 2). Very similar results were found by Marović and Senčar (1999) for absorbed dose rates from soils around the TENORM disposal site, where the highest contribution was from uranium series radionuclides (which includes ^{226}Ra), followed by thorium series radionuclides (^{232}Th) and ^{40}K, in decreasing order. It may be assumed that the pattern observed in Fig. 3 gives a typical representation of a typical karstic area, considering radionuclides contribution to radiological risks.

Conclusions

Radiological risks were the most pronounced in soils in which the highest D, D_{ef}, $\mathrm{Ra}_{\mathrm{eq}}$ and H_{ex} were determined compared to other types of studied samples. D and D_{ef} in the Kaštela Bay soils regularly exceeded average values for world soils. However, $\mathrm{Ra}_{\mathrm{eq}}$ and H_{ex} were below the recommended limits in all samples and it can be concluded that there are no adverse radiological effects in the researched area. ^{226}Ra was by far the most important contributor to D and H_{ex} in limestones s.l., while its contribution was significantly lower in other sample types. It was still the largest contributor in stream sediments as well, but only few percentages higher than ^{232}Th. ^{40}K was the largest contributor in marls and ^{232}Th in soils (with small differences between ^{232}Th and ^{226}Ra in soils). Although ^{40}K presented the highest massic activities of all studied radionuclides in all sample types, it was found that it contributed the least to D and H_{ex} in total in all samples. The greatest attention in terms of future radiological protection should be given to ^{226}Ra, especially when taking into account that its progeny is ^{222}Rn, which is one of the main natural sources of radiation exposure for humans.

Obtained results reflect typical karstic environment, coupled with flysch/marl influence. Therefore, Kaštela Bay coastal area may be considered a model karstic terrestrial environment comprising two different geological backgrounds, limestones s.l. and marls, and sediments developed on them.

Declarations

Ethics approval and consent to participate: Not applicable.

Consent for publication: Not applicable.

Availability of data and materials: The datasets used and analysed during the current study are available from the corresponding author on reasonable request.

Competing interests: The authors declare that they have no competing interests.

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Authors’ contributions: ILM – Conceptualization, methodology, collection of samples, data collection and analysis, interpretation of data, preparation and writing of the article; DB – Conceptualization and methodology of sampling, collection of samples. All authors read and approved the final manuscript.

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Figure captions

Fig. 1 Location and simplified lithological composition of the Kaštela Bay area with sampling locations on the coastal area (after Fritz 1994; Lovrenčić Mikelić et al. 2021; Marinčić et al. 1971)

Fig. 2 Relative contributions of ^{40}K, ^{226}Ra and ^{232}Th to outdoor absorbed dose rates in air and to external hazard indices in limestones s.l. (K), marls (L), stream sediments (PS) and soils (T)

Fig. 3 Relative total contributions of ^{40}K, ^{226}Ra and ^{232}Th to external hazard indices in all samples

Figure 1

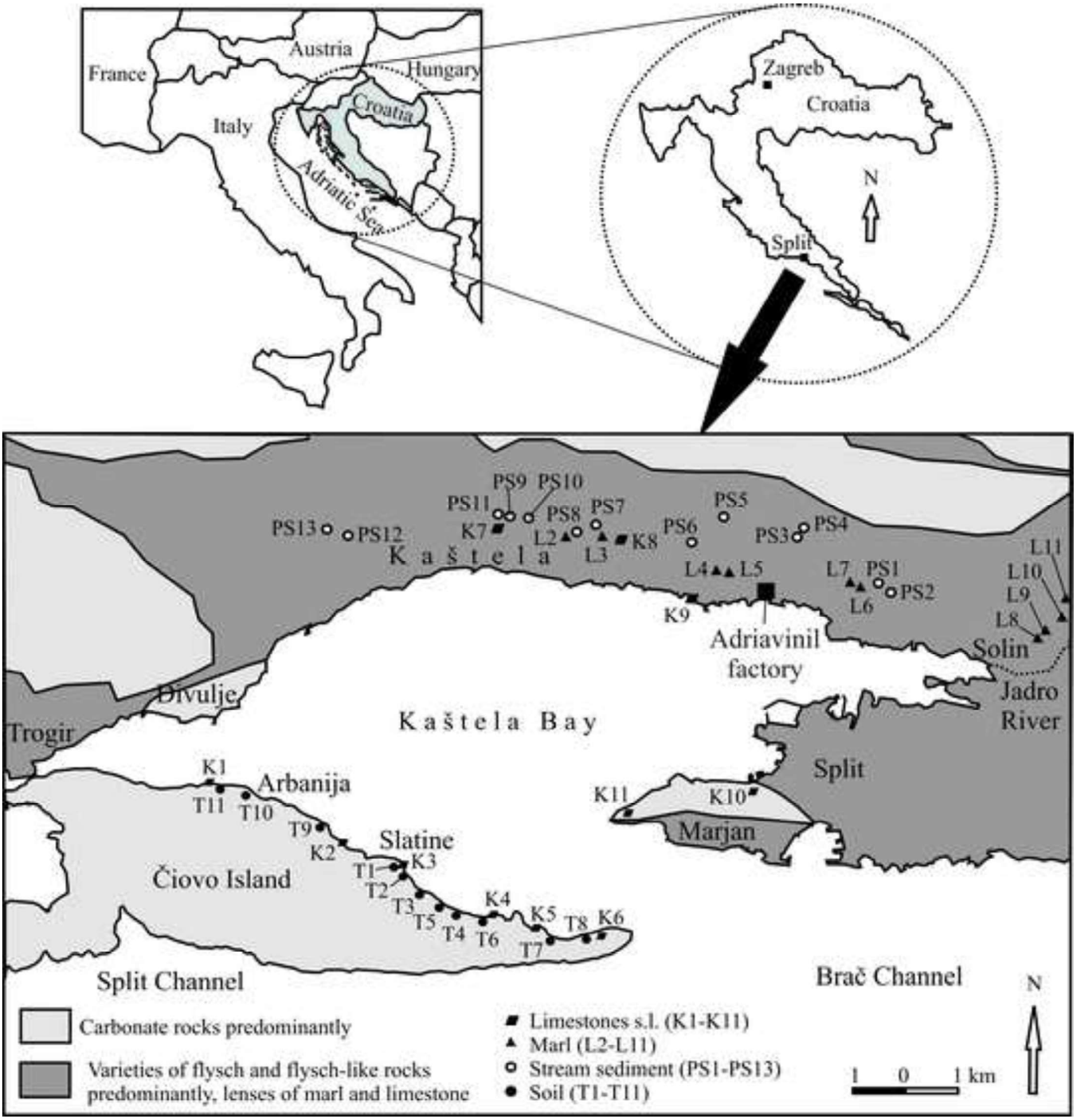

France
Austria
Hungary
Italy
Croatia
Adriatic Sea
Zagreb
Croatia
N
Split
PS9
PS10
PS11
PS13
PS12
K7
PS8
PS7
L2
L3
K8
PS6
PS5
PS3
PS4
Kaštela
L4
L5
L7
PS1
PS2
L6
L11
L10
L9
L8
K9
Adriavinil factory
Solin
Jadro River
Divulje
Trogir
Kaštela Bay
Split
K1
Arbanija
T11
T10
T9
K2
Slatine
K3
T1
T2
T3
T5
T4
T6
K4
K5
T8
K6
T7
Čiovo Island
K11
K10
Marjan
Split Channel
Brač Channel
N
Carbonate rocks predominantly
Varieties of flysch and flysch-like rocks predominantly, lenses of marl and limestone
Limestones s.l. (K1-K11)
Marl (L2-L11)
Stream sediment (PS1-PS13)
Soil (T1-T11)
1
0
1 km

Figure 2

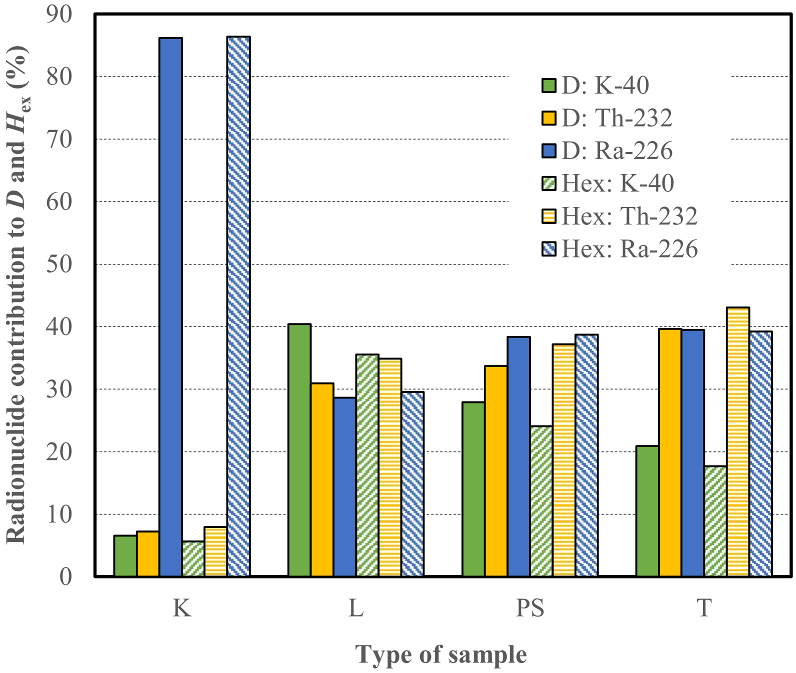
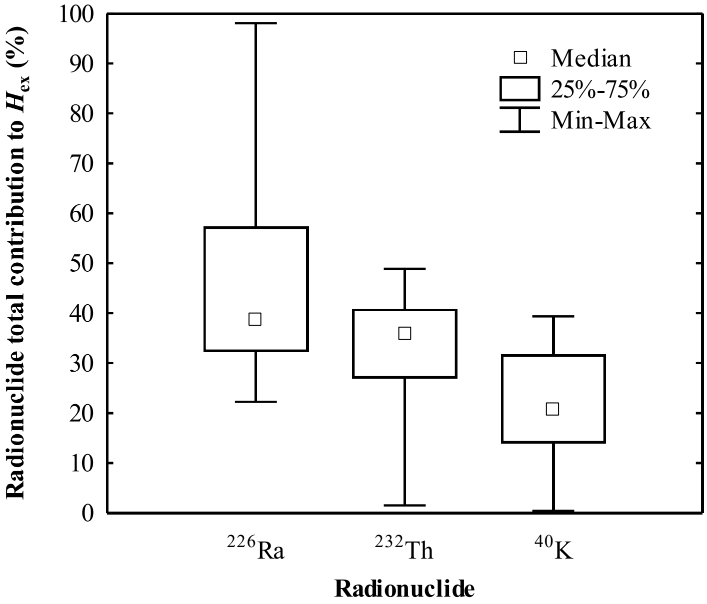

90
80
70
60
50
40
30
20
10
0
Radionuclide contribution to D and H_{ex} (%)
D: K-40
D: Th-232
D: Ra-226
Hex: K-40
Hex: Th-232
Hex: Ra-226
K
L
PS
T
Type of sample

Figure 3



Radionuclide total contribution to H_{ex} (%)
100
90
80
70
60
50
40
30
20
10
0
□ Median
25%-75%
Min-Max
^{226}Ra
^{232}Th
^{40}K
Radionuclide

Table 1

Table 1 Geographical coordinates of the Kaštela Bay coastal area sampling locations

Sample	Sampling location coordinates		Sample	Sampling location coordinates	
	N	E		N	E
K1	43°30'55.979"	16°17'45.249"	PS3	43°33'24.32"	16°26'7.881"
K2	43°30'17.631"	16°19'38.935"	PS4	43°33'29.907"	16°26'13.673"
K3	43°30'3.656"	16°20'30.225"	PS5	43°33'37.441"	16°25'5.934"
K4	43°29'32.402"	16°21'46.209"	PS6	43°33'21.59"	16°24'37.264"
K5	43°29'24.247"	16°22'22.772"	PS7	43°33'33.125"	16°23'16.528"
K6	43°29'18.434"	16°23'18.111"	PS8	43°33'28.689"	16°23'0.509"
K7	43°33'31.301"	16°21'53.006"	PS9	43°33'38.632"	16°22'3.784"
K8	43°33'23.861"	16°23'37.893"	PS10	43°33'37.678"	16°22'18.973"
K9	43°32'46.815"	16°24'37.24"	PS11	43°33'40.306"	16°21'52.855"
K10	43°30'47.176"	16°25'32.857"	PS12	43°33'27.915"	16°19'45.588"
K11	43°30'32.809"	16°23'38.475"	PS13	43°33'31.982"	16°19'27.637"
L2	43°33'25.208"	16°22'50.525"	T1	43°30'2.469"	16°20'21.717"
L3	43°33'25.345"	16°23'21.656"	T2	43°29'55.953"	16°20'28.994"
L4	43°33'3.578"	16°24'58.419"	T3	43°29'44.989"	16°20'42.533"
L5	43°33'2.414"	16°25'9.356"	T4	43°29'31.498"	16°21'14.034"
L6	43°32'52.516"	16°27'1.59"	T5	43°29'37.584"	16°20'59.683"
L7	43°32'55.461"	16°26'52.466"	T6	43°29'27.886"	16°21'36.567"
L8	43°32'19.461"	16°29'31.438"	T7	43°29'15.628"	16°22'33.529"
L9	43°32'24.521"	16°29'37.928"	T8	43°29'16.491"	16°23'4.72"
L10	43°32'32.813"	16°29'52.985"	T9	43°30'26.87"	16°19'18.639"
L11	43°32'43.834"	16°29'56.087"	T10	43°30'47.91"	16°18'16.181"
PS1	43°32'55.409"	16°27'16.868"	T11	43°30'51.249"	16°17'53.984"
PS2	43°32'49.102"	16°27'27.326"			

K – limestones s.l., L – marl, PS – stream sediment, T – soil

Table 2 Basic statistical parameters of ^{40}K, ^{226}Ra and ^{232}Th massic activities in rocks and sediments of the Kaštela Bay coastal area

Type of sample	Statistical parameter	^{40}K (Bq/kg)	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)
K (N = 11)	Range	0.5 – 69	9.4 – 60	0.1 – 3.4
	$\bar{x}$	14	19	1.0
	Median	7.2	14	0.8
	SD	19	15	0.9
L (N = 10)	Range	148 – 284	8.1 – 20	8.6 – 17
	$\bar{x}$	215	14	11
	Median	194	14	9.9
	SD	54	4.3	3.2
PS (N = 13)	Range	46 – 310	8.2 – 47	4.8 – 36
	$\bar{x}$	193	23	17
	Median	212	21	15
	SD	89	11	9.4
T (N = 11)	Range	168 – 581	28 – 198	20 – 84
	$\bar{x}$	463	85	62
	Median	518	72	71
	SD	140	48	20

K – limestones s.l., L – marl, PS – stream sediment, T – soil, N – number of samples, $\bar{x}$ – mean value, SD – standard deviation

Table 3

Table 3 Basic statistical parameters for radiological risks from radionuclides in rocks and sediments of the Kaštela Bay coastal area

Type of sample	**Statistical parameter**	***D* (nGy/h)**	**D_{ef} (mSv/y)**	**Ra_{eq} (Bq/kg)**	**H_{ex}**
K	Range	4.4 – 29	0.0054 – 0.035	9.6 – 62	0.026 – 0.17
	$\bar{x}$	10	0.012	22	0.058
	Median	8.0	0.010	17	0.047
	SD	6.7	0.0082	14	0.039
L	Range	15 – 31	0.019 – 0.038	32 – 66	0.086 – 0.18
	$\bar{x}$	22	0.027	47	0.13
	Median	20	0.024	41	0.11
	SD	5.8	0.0072	12	0.033
PS	Range	10 – 57	0.012 – 0.069	21 – 123	0.058 – 0.33
	$\bar{x}$	29	0.035	62	0.17
	Median	26	0.032	54	0.15
	SD	14	0.017	30	0.082
T	Range	32 – 158	0.039 – 0.19	70 – 346	0.19 – 0.93
	$\bar{x}$	96	0.12	209	0.56
	Median	96	0.12	210	0.57
	SD	36	0.044	78	0.21

D – outdoor absorbed dose rate in air, D_{ef} – annual outdoor effective dose rate, Ra_{eq} – radium equivalent activity, H_{ex} – external hazard index, K – limestones s.l., L – marl, PS – stream sediment, T – soil, $\bar{x}$ – mean value, SD – standard deviation