



# Uranium isotopes and several heavy elements in selected waters in Quang Nam-Da Nang provinces, Central Vietnam

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## Abstract

$^{238}\text{U}$  and  $^{234}\text{U}$  concentrations,  $^{238}\text{U}/^{234}\text{U}$  ratios and Na, Ca, Mg, K, Al, As, Cd concentrations were measured in selected surface waters (streams, rivers and lakes), ground waters (dug wells) and underground waters (drill wells and thermal waters) in Quang Nam and Da Nang provinces, Central Vietnam. The mineralization was  $< 500 \text{ mg L}^{-1}$  and Al, As, Cd contents were a few tenths of  $\mu\text{g L}^{-1}$ .  $^{234}\text{U}$  and  $^{238}\text{U}$  activities were between  $0.47\text{--}27.6 \text{ mBq L}^{-1}$  and  $0.6\text{--}15.0 \text{ mBq L}^{-1}$  respectively, these values are lower than WHO recommended limits. Uranium contents trended as  $U_{\text{rivers}} < U_{\text{streams}} < U_{\text{lakes}} < U_{\text{dig wells}} < U_{\text{thermal water}} < U_{\text{drill wells}}$ . The  $^{234}\text{U}/^{238}\text{U}$  ratio ranged from 0.69 to 2.31 with 1.26 on average. For groundwaters, the ratio scattered around one. Effects of Nong Son uranium deposit located in Quang Nam region were not observed.

**Keywords** Water reservoirs · Uranium isotopes · Heavy metals · Radioactivity · Vietnam

## Introduction

The hydrological cycle's heavy components' mobility, persistence, and potential toxicity can have a major effect on the environment. Emissions of heavy elements into the

atmosphere can originate from both natural and industrial sources such as: rain, snow, and dust dump into water reservoirs [1]. There are three natural uranium isotopes including  $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$ , and they are often present in trace amounts in natural waters [2].  $^{235}\text{U}$  abundance in uranium is approximately 0.7% and it can be considered constant in all natural environments, therefore in this paper only  $^{238}\text{U}$  and its progeny  $^{234}\text{U}$  are considered. The presence of uranium in water is mainly derived from dissolving minerals from rock formations [3–5] or releases from mining industry, nuclear fuel cycle, coal combustion, and use of phosphate fertilizers [6]. Many factors influence the uranium concentration in groundwater, including the content of uranium in the aquifer matrix, as well as current geochemical, hydrochemical and oxidation conditions [4, 7, 8]. Uranium can exist in different oxidation states in aqueous media [9]. In a mildly oxidizing environment, uranium exists with a +6 valence state, while with the reduction conditions, uranium occurs in the +4 valence state and gets removed from water into the solid phase [10, 11]. The direct recoil effect is a main phenomenon leading to the preferential mobilization of  $^{234}\text{U}$  over  $^{238}\text{U}$  [11–13]. The relation of uranium isotopes and their activity ratio ( $^{234}\text{U}/^{238}\text{U}$ ) for prevailing conditions in groundwater may offer new approaches to the models of

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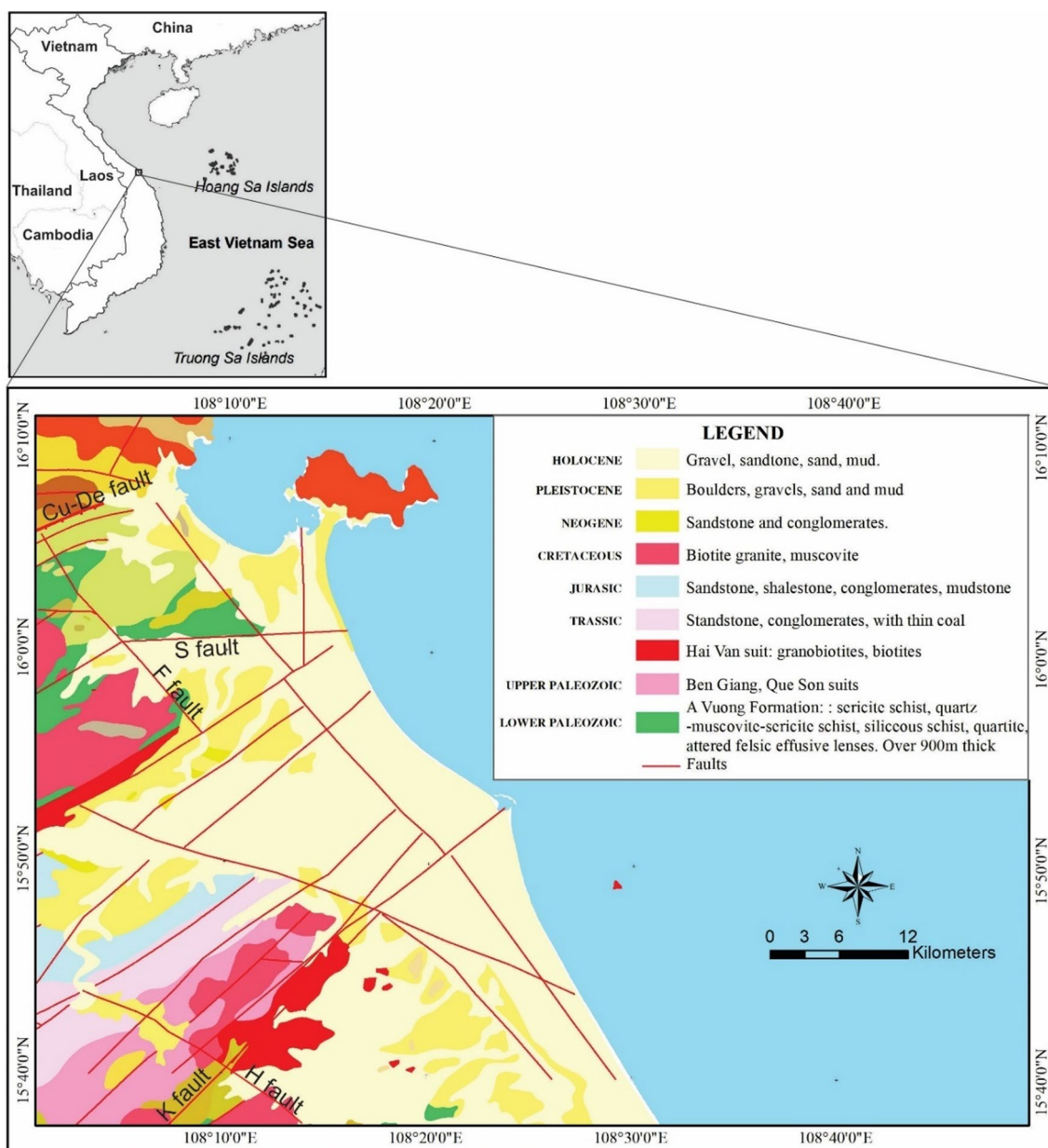
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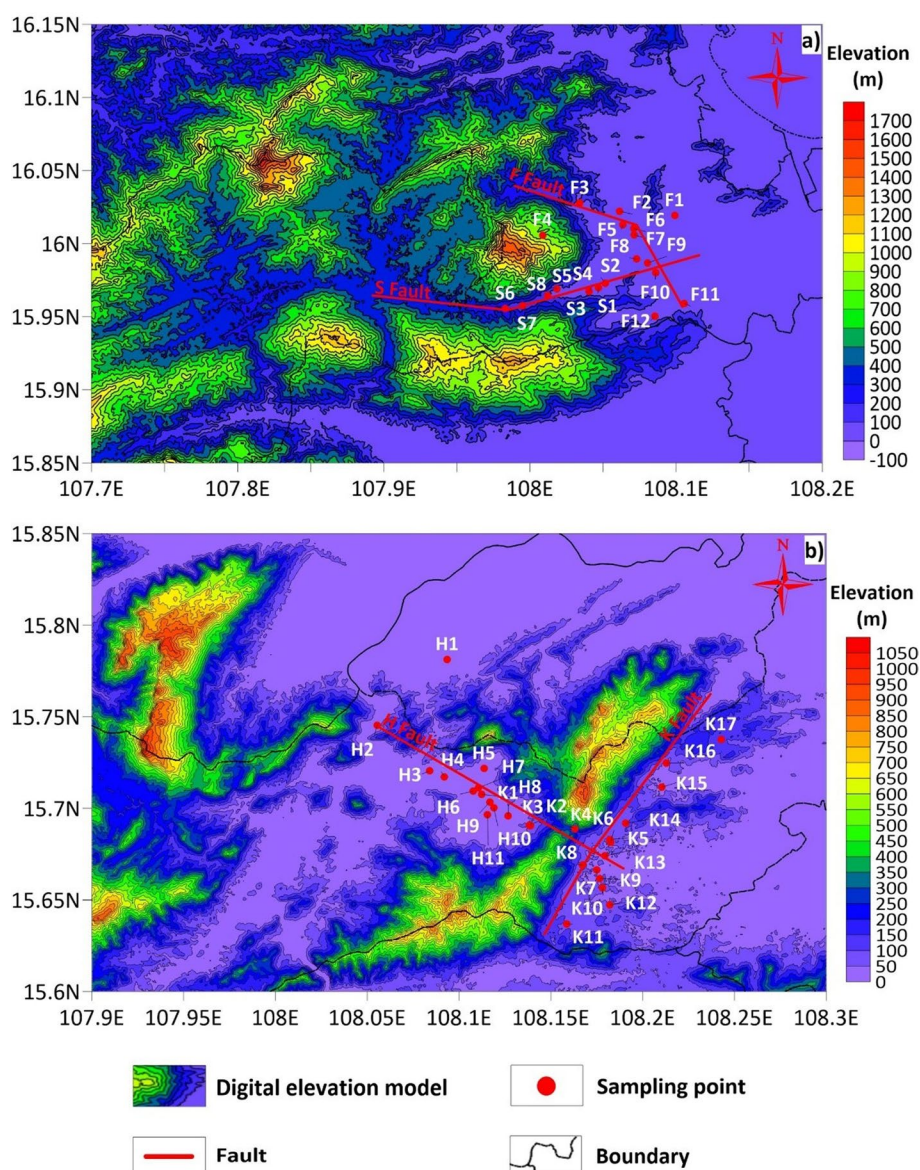


**Fig. 1** Localization of the Quang Nam-Da Nang regions in Vietnam and its geological sketch modified from National Agency for Science and Technology Information of Vietnam [32] and Duong et al. [33]

hydrogeological and hydrogeochemical processes [5]. As an inherent reference level for this system, the uranium isotope ratio in the undisturbed system was employed [11]. For example, the uranium ratio can be useful in characterizing the mixing zones [3, 12–14], as well as the process of interaction between water and rock, and describing the hydrological pathway [11, 15]. Uranium exhibits both chemical and radiological toxicities, this could significantly impact on the health of the aquatic organisms and humans using the water for drinking above a certain threshold. Uranium

isotopes are present in natural water as well, when consumed drinking water by humans, they can be associated with a cascade of health effects such as kidney disease and leukemia in humans upon drinking water rich in uranium [16, 17]. Uranium is excreted through the kidneys which in turn get damaged and accumulate in the skeleton due to the heavy metal properties of uranium [18, 19]. Toxicity of ingested uranium can vary over a broad range depending on multiple factors, including the chemical form, and acceptable

**Fig. 2** Sampling points shown on the morphological sketch of the study region



drinking water concentration values range from  $2 \mu\text{g L}^{-1}$  in Japan to  $1700 \mu\text{g L}^{-1}$  in Russia [18].

Hydrogeological research is an important issue due to exploitation and indiscriminate water use common in Vietnam, and the resulting risk of pollution in the surface water caused by human activities [20–22]. In the surface water of most regions in Far East countries, there are often high concentrations of several harmful elements, such as arsenic, cadmium, mercury [23]. In the Quang Nam–Da Nang region in the middle part of Vietnam, there are many faults, strong magmatic activity, and as a consequence there are different blocks of heated aquifer formations which could be a source of thermal water [24–26]. In the area neighboring the study region, there are some uranium deposits [27, 28], so the water could be contaminated with radioactive substances,

as the influence of mining was noticeable in case of North Vietnam [29].

The purposes of this study include (1) determining the  $^{234}\text{U}$  and  $^{238}\text{U}$  concentrations and their activity ratio ( $^{234}\text{U}/^{238}\text{U}$ ) in selected water sources; (2) performing the hydrological characterization of the studied water resources; (3) assessing the safety regarding the radiological effects on human health due to the consumption of water contaminated with uranium and heavy elements.

## Geological setting

The Quang Nam and Da Nang regions are located in the middle part of Vietnam. The region is characterized by the strong uplift of the Kon Tum–Da Lat massif and the deep

**Table 1** The selected chemical, physical parameters and concentrations of uranium isotopes in the water samples

Code	Water type	Depth (m)	pH	Eh (mV)	EC ( $\mu\text{S}/\text{cm}$ )	TDS (ppm)	T ( $^{\circ}\text{C}$ )	Uranium ( $\text{mBq L}^{-1}$ )		
								$^{234}\text{U}$	$^{238}\text{U}$	$^{234}\text{U}/^{238}\text{U}$
F1	Lake	–	7.62	17.3	65.6	71.1	27.8	$13.8 \pm 1.46$	$8.39 \pm 1.08$	$1.64 \pm 0.27$
F12	Lake	–	7.73	52.1	29.1	14.3	25.1	$7.15 \pm 0.72$	$5.49 \pm 0.62$	$1.30 \pm 0.20$
H1	Lake	–	7.49	106	38.9	20.2	26.7	$0.47 \pm 0.08$	$0.56 \pm 0.09$	$0.84 \pm 0.20$
H2	Lake	–	7.61	110	48.2	24.5	25.3	$2.50 \pm 0.25$	$3.05 \pm 0.27$	$0.82 \pm 0.11$
H5	Lake	–	7.68	102	85.6	44.3	30.5	$2.65 \pm 0.33$	$2.29 \pm 0.31$	$1.16 \pm 0.21$
K5	Lake	–	6.78	36.7	58.9	30.3	28.5	$6.54 \pm 0.63$	$5.41 \pm 0.56$	$1.21 \pm 0.17$
K8	Lake	–	8.92	71.2	74.4	37.8	31.8	$4.62 \pm 0.48$	$3.71 \pm 0.42$	$1.25 \pm 0.19$
K11	Lake	–	7.22	101	62.3	31.2	27.8	$4.30 \pm 0.41$	$3.68 \pm 0.35$	$1.17 \pm 0.16$
K16	Lake	–	7.46	75.8	60.2	30.1	29.0	$7.47 \pm 0.74$	$5.93 \pm 0.65$	$1.26 \pm 0.19$
min			6.78	17.3	29.1	14.3	25.1	0.47	0.56	0.82
max			8.92	110	85.6	71.1	31.8	13.8	8.39	1.64
avg			7.61	74.7	58.1	33.7	28.1	5.50	4.30	1.2
std			0.56	34	17	16	2.3	3.89	2.30	0.25
V [%]			7	45	30	49	8	71	50	21
F2	Stream	–	7.02	58.4	75.6	37.8	29.2	$1.44 \pm 0.24$	$0.99 \pm 0.20$	$1.45 \pm 0.38$
F4	Stream	–	7.03	57.8	76.4	38.1	29.4	$1.56 \pm 0.38$	$1.00 \pm 0.32$	$1.56 \pm 0.63$
F7	Stream	–	7.21	109	73.3	35.6	30.1	$2.30 \pm 0.07$	$1.56 \pm 0.05$	$1.47 \pm 0.06$
S4	Stream	–	7.08	103	26.2	13.2	30.1	$2.56 \pm 0.27$	$2.14 \pm 0.24$	$1.20 \pm 0.18$
S6	Stream	–	7.57	84.5	37.8	18.9	23.0	$2.20 \pm 0.19$	$1.58 \pm 0.18$	$1.39 \pm 0.20$
S7	Stream	–	7.41	103	28.9	14.3	26.1	$2.09 \pm 0.27$	$2.32 \pm 0.28$	$0.90 \pm 0.16$
S8	Stream	–	8.02	99.1	53.4	26.7	25.0	$4.49 \pm 0.45$	$3.82 \pm 0.41$	$1.18 \pm 0.17$
K2	stream	–	7.71	125	35.2	18.2	29.5	$3.71 \pm 0.67$	$3.12 \pm 0.34$	$1.19 \pm 0.25$
K4	Stream	–	7.56	90.3	33.1	16.7	29.1	$3.70 \pm 0.93$	$2.56 \pm 0.24$	$1.45 \pm 0.39$
K15	Stream	–	6.23	150	136	68.1	27.5	$6.89 \pm 1.65$	$7.59 \pm 0.66$	$0.91 \pm 0.23$
min			6.23	57.8	26.2	13.2	23.0	1.44	0.99	0.90
max			8.02	150	136	68.1	30.1	6.89	7.59	1.56
avg			7.2.8	98.0	57.6	28.7	27.9	3.09	2.67	1.27
std			0.49	27.9	33.9	16.9	2.44	1.66	1.95	0.23
V [%]			7	29	59	59	9	54	73	18
F9	River	–	7.43	92.1	44.1	23.2	27.4	$1.72 \pm 0.21$	$1.77 \pm 0.21$	$0.97 \pm 0.23$
S3	River	–	7.56	124	42.0	21.3	29.4	$1.10 \pm 0.51$	$0.80 \pm 0.29$	$1.38 \pm 0.23$
F3	Dug well	30	7.12	42.2	25.6	14.4	29.5	$3.32 \pm 0.42$	$3.76 \pm 0.45$	$0.88 \pm 0.15$
F5	Dug well	6	6.13	130	60.1	30.1	30.5	$2.56 \pm 0.07$	$2.10 \pm 0.05$	$1.22 \pm 0.04$
F10	Dug well	10	6.31	182	34.3	16.7	26.4	$11.9 \pm 1.08$	$12.9 \pm 1.14$	$0.92 \pm 0.12$
S1	Dug well	10	6.67	154	68.9	35.6	27.6	$5.61 \pm 0.67$	$4.12 \pm 0.51$	$1.36 \pm 0.23$
S2	Dug well	10	6.33	116	85.1	42.1	28.2	$8.94 \pm 1.04$	$6.23 \pm 1.80$	$1.43 \pm 0.45$
H7	Dug well	9	6.31	146	110	54.5	27.8	$3.72 \pm 0.43$	$1.97 \pm 0.30$	$1.89 \pm 0.36$
K7	Dug well	9	6.56	161	325	164	28.7	$1.43 \pm 0.21$	$1.34 \pm 0.21$	$1.07 \pm 0.23$
K14	Dug well	10	6.44	125	151	74.5	29.3	$5.05 \pm 0.45$	$2.81 \pm 0.33$	$1.80 \pm 0.27$
min			6.13	42.1	25.6	14.4	26.4	1.43	1.34	0.88
max			7.12	182	325	164	30.5	11.9	12.9	1.89
avg			6.48	132	108	53.9	28.5	5.32	4.40	1.32
std			0.31	42.1	96.7	48.6	1.28	3.50	3.76	0.38
V [%]			5	32	90	89	4	66	85	28
F6	Drilled wells	25	5.45	182	162	81.1	28.3	$0.91 \pm 0.17$	$1.01 \pm 0.18$	$0.90 \pm 0.23$
F8	Drilled wells	70	6.73	74.5	160	81.3	28.2	$3.80 \pm 0.1$	$3.15 \pm 0.09$	$1.21 \pm 0.05$
H3	Drilled wells	55	7.71	–40	640	317	28.4	$27.6 \pm 1.26$	$11.9 \pm 0.71$	$2.32 \pm 0.18$
H4	Drilled wells	50	6.13	153	209	103	27.5	$1.74 \pm 0.19$	$1.08 \pm 0.15$	$1.61 \pm 0.28$



**Table 1** (continued)

Code	Water type	Depth (m)	pH	Eh (mV)	EC ( $\mu\text{S}/\text{cm}$ )	TDS (ppm)	T ( $^{\circ}\text{C}$ )	Uranium ( $\text{mBq L}^{-1}$ )		
								$^{234}\text{U}$	$^{238}\text{U}$	$^{234}\text{U}/^{238}\text{U}$
H6	Drilled wells	70	6.45	150	208	104	27.4	$2.34 \pm 0.30$	$2.64 \pm 0.32$	$0.89 \pm 0.16$
H8	Drilled wells	60	9.45	− 182	400	200	29.0	$6.47 \pm 0.67$	$5.53 \pm 0.61$	$1.17 \pm 0.18$
H10	Drilled wells	18	6.14	37.4	235	119	30.1	$7.07 \pm 0.86$	$5.92 \pm 0.78$	$1.19 \pm 0.21$
H11	Drilled wells	28	6.03	87.1	83.1	41.1	27.6	$9.47 \pm 1.08$	$8.01 \pm 0.97$	$1.18 \pm 0.20$
K1	Drilled wells	18	6.12	135	110	55.6	28.7	$3.53 \pm 0.45$	$3.11 \pm 0.42$	$1.14 \pm 0.21$
K3	Drilled wells	7	6.89	45.6	166	86.7	29.0	$3.98 \pm 0.41$	$2.45 \pm 0.32$	$1.62 \pm 0.27$
K6	Drilled wells	16	5.78	166	65.6	33.2	30.2	$5.74 \pm 0.69$	$3.59 \pm 0.53$	$1.60 \pm 0.30$
K10	Drilled wells	50	6.32	86.2	214	102	32.1	$10.3 \pm 1.18$	$14.9 \pm 1.49$	$0.69 \pm 0.10$
K12	Drilled wells	55	7.31	79.1	275	137	28.7	$12.1 \pm 1.16$	$5.83 \pm 0.75$	$2.08 \pm 0.33$
K13	Drilled wells	15	6.24	142	143	73.1	29.8	$12.6 \pm 1.32$	$10.5 \pm 1.14$	$1.20 \pm 0.18$
K17	Drilled wells	30	8.22	44.4	33.3	15.6	30.0	$12.9 \pm 0.83$	$9.50 \pm 0.69$	$1.36 \pm 0.13$
min			5.45	− 182	33.3	15.6	27.4	0.91	1.01	0.69
max			9.45	182	640	317	32.1	27.6	14.9	2.32
avg			6.73	77.3	207	103	29.0	8.04	5.94	1.34
std			1.05	93.4	150	74.3	1.26	6.75	4.19	0.44
V [%]			16	121	72	72	4	84	71	33
F11	Thermal	50	9.21	− 202	638	318	41.0	$12 \pm 1.2$	$10.4 \pm 1.1$	$1.15 \pm 0.17$
S5	Thermal	50	8.43	97.8	900	450	38.0	$1.57 \pm 0.24$	$1.74 \pm 0.25$	$0.90 \pm 0.19$
H9	Thermal	unk <sup>1</sup>	9.42	− 251	481	236	55.0	$5.60 \pm 0.66$	$5.10 \pm 0.68$	$1.10 \pm 0.20$
K9	Thermal	unk	9.89	− 254	473	260	48.0	$1.28 \pm 0.2$	$1.28 \pm 0.2$	$1.00 \pm 0.22$
min			8.43	− 254	473	236	38.0	1.28	1.28	0.90
max			9.89	97.8	900	450	55.0	12	10.4	1.15
avg			9.23	− 152	623	316	45.5	5.11	4.63	1.04
std			0.61	168	199	95.7	7.59	5.00	4.21	0.11
V [%]			7	111	32	30	17	98	91	11

<sup>1</sup>-unk, unknown

subsidence of the continental shelf, creating mountainous terrain, hilly zones, plateaus, and plains where elevation gradually decreases from west to east. In the study area, there are many faults dividing the region onto the local sub-areas with different water surface reservoirs such as lakes, streams and rivers. The faults basically create two perpendicular fault systems, the first system has the sub-longitudes northeast-southwest trending directions, and the second-northwest-southeast (Fig. 1). Most areas of Quang Nam-Da Nang regions present geological formations from the Proterozoic to the Quaternary. In some places, there are several epoch formations missing [30]. Most of the Proterozoic and Paleozoic sedimentary formations are metamorphosed into claystone, with quartz, biotite, dolomite and marble. The Mesozoic formations are mainly composed of conglomerates, shale stone and sandstones with coarse granular structure through medium to fine-grained. The Cenozoic period is presented by the Neogene and Pleistocene sediments, which are characterized by boulders, gravel, sand, and mud with organic remains [31].

There are series of Permian and Triassic intrusive metamorphic suits such as A Vuong, Ben Giang, Que Son, and Hai Van [32]. The intrusive rocks are often metamorphosed and occur as grano-biotites, biotites, sericite schist, quartz-muscovite-sericite schist, and altered felsic effusive lenses. However, these formations are dense with very low porosity and quite water poor. A major water storage formation is the Holocene diluvial formation. The sand, mud, and gravel-filled river deltas that make up the Holocene diluvial deposits. These formations range in extent from several tens to hundreds of square kilometers, and their thickness extends from a few meters to a dozen meters [33]. The Pleistocene water hosting formation is distributed in all the regions, with a thickness from 7 to 15 m. The main components of Pleistocene sediments include tiny grain sands with quartz, gravel, boulders, and mud. These formations are often poor in natural radionuclides. Finally, the water hosting Neogene sediments in Quang Nam province consist of mudstone, sandstone with gravel, conglomerates, and organic materials, having a thickness from 50 to 120 m [33].

## Methods and materials

### Sampling

During summer 2022, the sampling was performed along the four major faults in the study area, where there are strongly fissured aquifers and dense population. The faults are located along the four lines F, S, H and K. The F and S lines are in Hoa Vang district in the Da Nang province (Fig. 2a), and H and K lines are in the Que Son and Nong Son districts in Quang Nam province (Fig. 2b). A total of 48 samples were collected from streams, rivers and lakes as surface waters, dug wells as ground water and drill wells and thermal waters as underground waters. The volume of each sample was approximately 10 L. The water samples were filtered, and the pH of the samples was reduced to 2 by adding nitric acid to preserve the samples and prevent precipitation and then they were transported to the laboratory for analysis.

### Method

#### Determination of the physical and chemical parameters

The pH, EC ( $\mu\text{S cm}^{-1}$ ), temperature ( $^{\circ}\text{C}$ ) and Eh (mV) of the waters were measured in-situ using Hanna instruments model HI2003-02<sup>TM</sup> and HI8314<sup>TM</sup>. The data of the mentioned parameters are presented in Table 1. The contents of the chemical elements in the water samples were determined using the inductively coupled plasma–optical emission spectrometry (ICP-OES) technique using an Optima 7300 DV spectrometer of the Perkin Elmer<sup>TM</sup>. Multi- and single element standard solutions from Sigma Aldrich® were used for the preparation of calibration curves. For every analyzed element: Ca, Mg, Na, K, Al and Cd, five standard samples with 0, 100, 250, 500 and 1000  $\mu\text{g L}^{-1}$  of the adequate element were prepared and measured, then the received results were used to build the calibration curves. For arsenic (As) the standard samples with 0, 10, 100 and 500  $\mu\text{g L}^{-1}$  were prepared. The concentration of the element in the analyzed sample was determined based on the measured count rate of the sample and the calibration curve of the element. The analytical procedure was performed according to the ISO 11885 standard method. For the verification of the method a weighted amount of standard solution was added to the analyzed sample. The coefficient of recovery is calculated by the following formula:

$$\alpha = \frac{S_m - S_x}{S_{st}} \quad (1)$$

where:  $\alpha$  is the coefficient of recovery,  $S_m$  [ $\mu\text{g L}^{-1}$ ] is the concentration of the analyzed sample with a known added

amount of standard solution,  $S_x$  [ $\mu\text{g L}^{-1}$ ] concentration is the analyzed element in the measured sample and  $S_{st}$  [ $\mu\text{g L}^{-1}$ ] is the concentration of the standard solution. The analyzed method was accepted if the coefficient of recovery should be higher than 80%.

The limit of detection (LOD) depended on the element, and for the elements tested in the study, it ranges in the order of a few hundredths of micrograms per liter to half milligram per liter. The concentration values of the measured main metal components and heavy elements in the water samples are shown in Table 2.

#### Determination of uranium isotopes

The determination of uranium isotopes in water samples were carried out following the procedure developed by Chau and his co-workers [35], 100 mBq aliquots of  $^{232}\text{U}$  tracer was added to five liters of each sample. The uranium in the 5-L water subsample was precipitated as ammonium uranium oxide  $(\text{NH}_4)_2\text{U}_2\text{O}_7$  with manganese dioxide ( $\text{MnO}_2$ ). To eliminate the other actinide elements such as thorium or actinium, the obtained precipitate was dissolved in 9 M HCl and passed through a Dowex1  $\times$  8 100–200 mesh chromatographic column. The uranium was eluted by adding 1 M HCl solution into column, then the solution was evaporated to dryness and the remains dissolved again in 1 M HCl. The uranium was micro-precipitated again by adding Mohr salt, neodymium chloride and HF acid. The precipitate was recovered using a membrane filter of 0.1  $\mu\text{m}$  pore size. After air drying at room temperature for at least 24 h, the sample was measured using an alpha spectrometer Alpha-Ensemble-4 ALPHA-DUOM1<sup>TM</sup>-coupled with a 450  $\text{mm}^2$  area Passivated Implanted Planar Silicon detector (PIPSD). The measurement time for each sample was set for the uncertainty of the count rate in the  $^{232}\text{U}$  peak to be less than 2%. The background count rate value generated from the chemical reagents, electronic noise, or alpha particles emitted from the surroundings was determined using a blank sample, which was prepared from distilled water and the background value was subtracted from the result of each water sample. Based on a comparison of the count rates under the peaks of the alpha particles with energies of 4.20 and 4.77 MeV emitted from  $^{238}\text{U}$  and from  $^{234}\text{U}$  respectively, with the count rate under the peak of alpha particles with an energy of 5.4 MeV emitted from the  $^{232}\text{U}$  tracer and its known activity added into the water sample at the very beginning of the chemical preparation, the concentrations of  $^{238}\text{U}$  and  $^{234}\text{U}$  in the water samples were determined. The average radiochemical recovery for uranium isotopes is greater than 50% and MDA is 0.5 mBqL<sup>-1</sup>.

## Results

### Physical and chemical characteristics

The chemical, physical parameters and uranium isotope concentrations of the water from the studied water sources are shown in Table 1. The surface water samples were from slightly acidic to alkaline, for lake and stream water the pH ranged from 6.23 to 8.92, while for the dug well waters the pH ranged from 6.13 to 7.12, the underground waters (drilled well) waters ranged from 5.45 to 9.45 while thermal waters were alkaline with pH values from 8.44 to 9.89. The Eh of the surface waters was from 17.3 mV to 150 mV and for dug well waters from 25.6 to 325 mV, for drilled well waters from – 182 to 182 mV and for thermal waters from – 254 to 97.8 mV. The mineralization (Total Dissolved solids-TDS) of surface waters was in the range of 13.2–71.1 mg/L, for dug well waters from 14.4–164 mg/L, for drilled well waters from 15.6–317 mg/L and for thermal waters from 236–450 mg/L.

### The chemical composition of the collected water samples

The concentrations of the analyzed main chemical components and several heavy metals of the studied waters are shown in Table 2. Generally, the mineralization of the waters is very low, below 500 mg L<sup>-1</sup>, and they can be classified as fresh water, but the clear increasing tendency of the mineralization and the main chemical components is observed from the surface waters through the groundwater to the underground waters. The ranges of Na, Ca, Mg and K concentrations are similar and range from a few mg L<sup>-1</sup> to several tens mg L<sup>-1</sup> (See appendix 1, Table 2), but in all of the studied water samples, the sodium concentration is higher than that of calcium, magnesium and potassium. The average concentrations of arsenic and aluminum are 0.01 and 47.6 µg L<sup>-1</sup>, respectively. These values are much lower than the maximum permissible concentrations in drinking water recommended by WHO, which are 10 µg L<sup>-1</sup> for As, and 900 µg L<sup>-1</sup> for Al [34]. There was no cadmium observed in the water sources, with the exception of lakes, in which cadmium ranges from below the limit of detection (LOD) up to 0.040 µg L<sup>-1</sup> with a mean concentration of 0.01 µg L<sup>-1</sup>. This value is far lower than the maximum safety value of 3 µg L<sup>-1</sup> recommended by WHO [34].

### Uranium in different water sources

The measured concentrations and their standard deviations of the uranium isotopes in the water samples are

presented in Table 1. The activity concentrations range from 0.56–14.9 mBq L<sup>-1</sup> with an average of 4.39 mBq L<sup>-1</sup> for <sup>238</sup>U, and from 0.47–27.6 with an average of 5.56 mBq L<sup>-1</sup> for <sup>234</sup>U (Table 1). The <sup>234</sup>U and <sup>238</sup>U average activity concentrations in groundwater and surface waters are 6.71 and 5.22 mBq L<sup>-1</sup>, and 3.97 and 3.23 mBq L<sup>-1</sup>, respectively.

The measured average activity concentrations of <sup>234</sup>U and <sup>238</sup>U are 8.04 and 5.94 mBq L<sup>-1</sup> for drilled wells, 5.12 and 4.63 mBq L<sup>-1</sup> for thermal water, and 5.32 and 4.40 mBq L<sup>-1</sup> for water from dug wells respectively. In the surface waters, the average activity concentrations of <sup>234</sup>U and <sup>238</sup>U are 5.50 and 4.30 mBq L<sup>-1</sup> for lakes; 1.41 and 1.29 mBq L<sup>-1</sup> for rivers; 3.1 and 2.67 mBq L<sup>-1</sup> for streams. The measured activities of <sup>234</sup>U and <sup>238</sup>U of all the studied waters are lower than the maximum permitted level in drinking water recommended by WHO, which is 1 Bq L<sup>-1</sup> and 10 Bq L<sup>-1</sup>, respectively [34], the recommendation for the maximum permissible chemical concentration of uranium is 30 µg L<sup>-1</sup> [18] which converts to a slightly stricter value of 758.4 mBq L<sup>-1</sup> <sup>238</sup>U (using the conversion factor 0.02528 Bq L<sup>-1</sup> per 1 µg L<sup>-1</sup> provided by [34]). It should be noted that the samples of the current study were taken in the rainy season, which might have caused a dilution effect. To put the data in context, the activity concentrations of <sup>234</sup>U and <sup>238</sup>U in the studied thermal waters are far lower than that in the thermal waters in North Vietnam, reported to have 197 mBq L<sup>-1</sup> for <sup>234</sup>U and 142 mBq L<sup>-1</sup> for <sup>238</sup>U on average [35]. Well waters measured previously in the vicinity of rare earth element and uranium mines in North Vietnam also showed orders of magnitude higher uranium concentrations, ranging from 38 mBq L<sup>-1</sup> to 5320 mBq L<sup>-1</sup> [29]. Heavily contaminated groundwater near the Dniprovske tailing storage facility in Ukraine can reach concentrations of 3200 to 92,900 mBq L<sup>-1</sup> [36]. In case of surface waters in Kazakhstan, uranium activity concentrations between 15.2 mBq L<sup>-1</sup> and 1567 mBq L<sup>-1</sup> (converted from 0.6 to 62 µg U L<sup>-1</sup>) were reported for a creek influenced by outflow from uranium mining adits [37]. Other mining activities might also lead to uranium mobilization, for example in the San Joaquin Valley the bicarbonate-rich produced waters caused median uranium concentrations up to 12,463 mBq L<sup>-1</sup> (493 µg L<sup>-1</sup>) in groundwater by mobilizing uranium. In comparison, our observations do not seem to show any significant influence from the uranium deposits identified in the area [20, 27].

### <sup>234</sup>U / <sup>238</sup>U ratio

The uranium activity ratio (<sup>234</sup>U/<sup>238</sup>U) ranges from 0.69 to 2.31, with an average of 1.26 (Table 1). In general, most of the <sup>234</sup>U/<sup>238</sup>U activity ratio (AR) values in the studied waters are comparable with the world average value [38].

Due to the dissolution, desorption and recoil effect i.e. the physical ejection of a recoil nucleus following alpha decay of its parent and geochemical conditions occurring in the water aquifer, there is often disequilibrium between uranium isotopes and the AR is usually higher than one [14, 39–42].

## Discussion

The measured chemical components of the collected water samples are shown in Table 1 and Table 2. The mineralization in the lake, stream and river waters is very low in the area, but mineralization as well as the concentrations of Na, Ca and Mg are lowest for river water, higher for stream water and highest for lake water, the phenomena can be related to the mobility of the water in the type of reservoirs. The water refreshment rate is highest in the river and the lowest in the lake, while residence time is the reverse, so lake water can reach higher concentration. Agricultural activity might also have some influence, phosphate fertilizers may contain soluble forms of uranium [43] and heavy elements, such as cadmium [44]. Sodium has the highest concentration in comparison with Ca, Mg and K, this feature is characteristic for the waters in the studied region. According to the hydrogeological rules, the mineralization and the concentration of the principal metals (Ca, Mg, Na and K) are highest in the thermal waters and lowest in the surface water. The concentration of heavy elements such as Al, As and Cd are far lower than the permissible upper limits recommended by WHO [34] and United States Environmental Protection Agency (US EPA) in drinking water, which is 0.01 ppm for As, 0.005 mg L<sup>-1</sup> for Cd; indicating the studied waters are not contaminated with the heavy elements. The variety (ratio of standard deviation to average value) of the concentration of the chemical components for each water type is high (from 30% up to over a hundred percent). Such a large variety could be a consequence of: (i) the mosaic characteristics of the morphology and geology in the study region; (ii) the very low concentrations of the elements in the water leading to the high standard deviation of the analyzed results; (iii) the variation values of the pH and Eh indicate that the discrepancy of the conditions prevailing in the surface water reservoirs are significantly smaller than that in the ground and underground water resources. The difference is one of the factors influencing the intervals variation of the chemical composition and uranium isotopes in the waters.

There is no clear relationship between EC and depth ( $r=0.54$ ), depth and TDS ( $r=0.52$ ) as well as between Eh and EC ( $r=-0.59$ ), Eh and TDS ( $r=-0.56$ ). Kindly check and confirm References are renumbered. References are true number

The oxidation–reduction potentials are the main factors controlling uranium mobilization in the aquatic environment [5, 13–15]. Excluding thermal waters, the observed Eh values in

most of the study waters are positive, beneficial for the formation of uranium compounds, where uranium has valence of +6 and is easily dissolved in such water, but the measured uranium concentrations in the waters are very low. This phenomenon could be related to: (i) low amount of uranium minerals in the host water formation; or (ii) the presence of the colloids leading to the immobilization of U(VI) [45, 46] or (iii) dilution by low activity water. Four thermal waters and two drilled well waters have strong negative Eh indicating redox conditions where the valence of uranium is 4+, in this case uranium is in immobile chemical forms and its activity in the waters should be expected to be relatively low.

Geology, hydrogeology and climate are also factors affecting the presence of uranium in water [11, 47]. The activity concentration of uranium in groundwater is related to its amount in the matrix aquifer [4]. High uranium content is commonly found in uranium-rich rocks such as granite or shale [2, 7, 8, 13]. The aquifers in the study regions are probably separated from the uranium deposits or only a small amount of uranium is leached from the uranium-rich rocks because of geological factors or related to climate factors. The sampling was carried out in September, which is in the annual rainy season. The rain is often heavy and lasts a few days, and in consequence, there are often floods [48, 49]. The large volume of water can dilute uranium in the waters. This fact is in agreement with the low water mineralization.

Ground and surface water are often characterized by both of uranium concentration and <sup>234</sup>U/<sup>238</sup>U activity ratio, which is commonly referred to as the uranium isotopic signature [50]. Selective leaching processes result in preferential dissolution and transport of <sup>234</sup>U when aquatic systems come into contact with minerals, improving the <sup>234</sup>U/<sup>238</sup>U ratio [51]. The <sup>234</sup>U/<sup>238</sup>U ratio is often characteristic of the water source, since this ratio is often related to the geology, hydrogeology and physical geochemical conditions prevailing in the water host formation. For uranium rich formations, the uranium activity ratio in water is often scattered around one, while in poor uranium formations, the ratio usually is significantly higher [38]. In the oxidizing environment, both uranium isotopes (<sup>234</sup>U, <sup>238</sup>U) have the same probability to enter the water, and as a consequence, their activity ratio is around one. The ratio of uranium activity isotopes observed in the surface waters (streams, rivers and lakes) were similar and the average value was near 1.23. In the dug well waters and drilled well waters the average ratio was 1.33. This value is a bit higher than that observed in the surface waters, possibly related with the redox potential (Eh) in underground conditions in comparison with that in the surface water. In the thermal waters the uranium isotope activity ratio is near one, probably related to the significantly higher interaction between the water and the host rocks due to high temperature and larger residence time.



**Table 2** Principal chemical composition and several harmful elements in the studied water

Code	Water type	Ca [mg L <sup>-1</sup> ]	Mg [mg L <sup>-1</sup> ]	Na [mg L <sup>-1</sup> ]	K [mg L <sup>-1</sup> ]	Al [μg L <sup>-1</sup> ]	As [μg L <sup>-1</sup> ]	Cd [μg L <sup>-1</sup> ]
LOD		0.1	0.02	0.06	0.03	0.02	0.02	0.003
F1	Lake	3.10	1.55	5.16	1.79	0.52	0.63	0.004
F12	Lake	1.99	0.32	4.08	1.03	8.30	0.50	0.008
H1	Lake	1.07	0.76	2.94	0.81	1.72	0.90	<LOD
H2	Lake	1.96	1.29	2.19	1.55	632	4.02	0.022
H5	Lake	3.42	1.27	2.83	0.44	1.74	1.43	0.009
K5	Lake	2.00	0.99	5.13	1.25	20.9	0.27	0.040
K8	Lake	3.99	1.41	5.53	0.81	2.90	0.90	<LOD
K11	Lake	2.03	1.01	5.06	1.15	24.3	0.10	0.021
K16	Lake	2.56	0.37	3.72	1.46	7.00	0.28	0.010
min		1.07	0.32	2.19	0.44	1.0	0.27	<LOD
max		3.99	1.55	5.53	1.79	632	4.02	0.040
avg		2.46	0.99	4.07	1.14	78.0	1.11	0.013
std		0.89	0.44	1.22	0.42	208	1.16	0.012
V[%]		37	44	30	37	268	104	92
F2	Stream	3.50	2.22	5.81	1.46	1.0	1.0	<LOD
F4	Stream	2.44	0.35	2.85	1.32	218	<LOD	<LOD
F7	Stream	3.07	2.09	5.25	1.29	2.0	<LOD	<LOD
S4	Stream	2.09	0.36	3.46	0.98	12.0	<LOD	<LOD
S6	Stream	3.16	0.44	3.44	1.20	6.0	2.0	<LOD
S7	Stream	2.79	0.46	3.29	0.84	14.0	1.0	<LOD
S8	Stream	4.17	0.49	3.41	1.31	18.0	1.0	<LOD
K2	Stream	2.75	0.58	4.19	0.79	3.0	<LOD	<LOD
K4	Stream	0.76	0.23	4.44	0.92	2.0	<LOD	<LOD
K15	Stream	2.76	0.87	4.71	2.19	32.0	<LOD	<LOD
min		0.76	0.23	2.85	0.79	1.0	<LOD	
max		4.17	2.22	5.81	2.19	218	2.0	
avg		2.75	0.81	4.08	1.23	31.0	1.0	
std		0.90	0.73	0.96	0.41	66.0	0.5	
V[%]		33	90	23	33	215	82	
F9	River	3.09	0.76	3.40	1.56	4.0	1.0	<LOD
S3	River	2.84	0.63	4.02	1.32	14.0	1.0	<LOD
F3	Dug well	2.19	0.34	3.22	1.41	8.0	<LOD	<LOD
F5	Dug well	2.45	1.87	5.14	1.73	1.0	<LOD	<LOD
F10	Dug well	1.98	0.12	3.63	1.03	2.0	2.0	<LOD
S1	Dug well	3.05	1.11	5.91	1.17	<LOD	<LOD	<LOD
S2	Dug well	4.54	1.37	4.73	1.07	<LOD	<LOD	<LOD
H7	Dug well	2.70	1.28	4.33	0.70	1.0	<LOD	<LOD
K7	Dug well	19.6	6.41	24.9	3.42	<LOD	<LOD	<LOD
K14	Dug well	4.36	2.12	9.10	0.82	5.0	<LOD	
min		1.98	0.12	3.22	0.70	nd	nd	
max		19.6	6.41	24.9	3.42	8.0	2.0	
avg		5.11	1.84	7.63	1.42	2.0	2.0	
std		5.93	1.97	7.23	0.87	3.0	0.5	
V [%]		116	108	95	61.5	117	25	
F6	Drilled wells	8.34	2.86	14.5	3.68	105	<LOD	<LOD
F8	Drilled wells	10.5	4.20	5.88	4.03	<LOD	<LOD	<LOD
H3	Drilled wells	36.7	9.45	34.6	1.08	2.0	4.0	<LOD
H4	Drilled wells	4.84	3.91	5.75	2.77	1.0	<LOD	<LOD
H6	Drilled wells	5.96	2.42	6.91	1.60	<LOD	<LOD	<LOD

**Table 2** (continued)

Code	Water type	Ca [mg L <sup>-1</sup> ]	Mg [mg L <sup>-1</sup> ]	Na [mg L <sup>-1</sup> ]	K [mg L <sup>-1</sup> ]	Al [μg L <sup>-1</sup> ]	As [μg L <sup>-1</sup> ]	Cd [μg L <sup>-1</sup> ]
H8	Drilled wells	2.43	0.05	73.8	1.09	9.0	<LOD	<LOD
H10	Drilled wells	10.8	3.99	22.4	6.76	<LOD	<LOD	<LOD
H11	Drilled wells	1.46	0.68	2.17	1.45	<LOD	<LOD	<LOD
K1	Drilled wells	5.26	2.13	8.35	4.08	<LOD	<LOD	<LOD
K3	Drilled wells	10.8	2.90	11.8	1.12	<LOD	<LOD	<LOD
K6	Drilled wells	2.78	0.59	7.84	2.83	<LOD	<LOD	<LOD
K10	Drilled wells	10.8	5.75	15.3	4.54	<LOD	<LOD	<LOD
K12	Drilled wells	10.3	7.07	11.1	2.16	<LOD	<LOD	<LOD
K13	Drilled wells	3.45	3.40	9.65	3.78	<LOD	<LOD	<LOD
K17	Drilled wells	18.7	5.33	11.5	1.84	450	<LOD	<LOD
min		1.46	0.05	2.17	1.08	<LOD	<LOD	<LOD
max		36.7	9.45	73.8	6.76	450	4.0	<LOD
avg		9.55	3.65	16.1	2.86	104	nd	nd
std		8.79	2.52	17.8	1.62	nd	nd	nd
V [%]		24	26.7	24.1	24	nd	nd	nd
F11	Thermal	0.46	0.03	98.2	3.13	4.0	1.0	<LOD
S5	Thermal	40.9	1.22	99.3	12.3	3.0	1.0	<LOD
H9	Thermal	1.54	0.01	73.9	2.85	25.0	8.0	<LOD
K9	Thermal	2.40	0.03	68.2	1.38	23.0	<LOD	<LOD
min		1.54	0.01	68.2	1.38	3.0	<LOD	nd
max		40.9	1.22	99.3	1.23	25.0	8.0	nd
avg		12.4	0.32	84.9	4.92	14.0	3.0	nd
std		19.1	0.59	16.2	4.98	12.0	3.0	nd
V [%]		154	186	19	101	86	100	nd

LOD, lower than limit of detection; nd, not determined

The annual effective dose from absorption of uranium isotopes in water can be calculated by formula as follows:

$$D = V \cdot (A_{U-238} \cdot e_{g-U238} + A_{U-234} \cdot e_{g-U234}) \quad (2)$$

where:  $D$  is the annual effective dose (mSv/year),  $A_{U-238}$  and  $A_{U-234}$  are the activity concentrations of  $^{238}\text{U}$  and  $^{234}\text{U}$  in the drinking water (mBq L<sup>-1</sup>),  $e_{g-U238}$  and  $e_{g-U234}$  are the effective dose resulting from the absorption of one Becquerel of  $^{238}\text{U}$  and  $^{234}\text{U}$  by humans at a given age respectively; while  $V$  is the volume of water consumed per year. The values of  $e_{g-U238}$  and  $e_{g-U234}$  were taken from International Basic Safety Standard [51].

The annual effective dose for adults (> 17 years old) and for children of 1–2 years old was calculated to be 0.0015 mSv/year and 0.0039 mSv/year, assuming the maximum concentrations of the uranium isotopes in the water from Table 1,  $^{238}\text{U} = 14.9 \text{ mBq L}^{-1}$  and  $^{234}\text{U} = 27.6 \text{ mBq L}^{-1}$  respectively, and an annual water consumption rate of 730 dm<sup>3</sup> (two liters per day). These values are far lower than the maximum permissible value (0.1 mSv/year) recommended by WHO [34].

## Conclusions

Forty-eight water samples from the Quang Nam and Da Nang regions in Vietnam were analyzed to determine the chemical concentrations of multiple elements and  $^{234}\text{U}$  and  $^{238}\text{U}$  as well as the ratio of  $^{234}\text{U}/^{238}\text{U}$  including surface water (rivers, streams, lakes) and groundwater (dug wells) and underground waters (borehole/drilled well water and thermal water). The observed uranium concentrations ranged from 0.6 to 14.9 mBq L<sup>-1</sup> with an average value of 4.39 mBq L<sup>-1</sup> for  $^{238}\text{U}$ , and 0.47 to 27.6 with an average value of 5.56 mBq L<sup>-1</sup> for  $^{234}\text{U}$ . In general, uranium concentrations are below the maximum level recommended by the WHO [34], so they can be considered safe in this regard. The trend of uranium activity concentrations in the studied water sources was:  $U_{\text{rivers}} < U_{\text{streams}} < U_{\text{lakes}} < U_{\text{dug wells}} < U_{\text{thermal water}} < U_{\text{drilled wells}}$ . The measured Eh data shows that the oxidizing conditions dominate in surface and ground waters, while in several underground waters (drill hole and thermal waters) reductive conditions are prevailing. The low uranium concentration

in the waters can be related to the water hosting sand and gravel Quaternary formations with low uranium and thorium concentrations and climate conditions in the region, but this should be verified by future investigations. Although there is a uranium deposit close to the studied regions, the uranium contents in the waters were observed as low, which could be related to the mosaic geological character of the studied region.

The observed  $^{234}\text{U}/^{238}\text{U}$  ratio ranges from 0.69 to 2.31. The average uranium isotope activity ratio was 1.2 and 1.27 for lake and steam waters respectively (surface water), for dug well water it was 1.32, 1.34 was observed for drilled well waters, but for thermal water was 1.04 (Table 1). These values probably are characteristic for the types of the studied waters, but this supposition demands deeper investigation in further studies. The uranium activity ratio in the thermal waters is around one, confirming the significantly larger interaction between thermal water with the host formation. This is also confirmed by the thermal waters having the large mineralization.

The mineralization of all studied waters was below  $500 \text{ mgL}^{-1}$  and the concentrations of heavy elements such as As, and Al are below the permissible maximum levels for drinking water. The Cd is present in measurable concentrations only in lake water, but these values are also far lower than that recommended maximum values by the US EPA. No public health issues were identified in this study. Uranium concentrations followed the same tendency as mineralization (TDS) in the observed samples.

## Appendix 1

See Table 2.

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## Declarations

**Conflict of interest** The authors declare that they have no financial or nonfinancial interests in the subject matter or materials addressed in this article.

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