



Gross alpha/beta activity concentrations in spa and mineral waters in North Vietnam

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Abstract

There is little available information on the radioactivity concentrations in the thermal and drinking water sources in North Vietnam. In the current study, the gross alpha and gross beta activity concentrations were determined in 8 water sources in the area. The average activities of gross alpha and beta in the 8 sources are 38.7 mBq L⁻¹ and 88.0 mBq L⁻¹. These activity concentrations are lower than WHO recommendations for drinking water. In this study, the gross alpha and gross beta show a weak correlation.

Keywords Gross alpha · Gross beta · Hot spring · North Vietnam

Introduction

The water has an indispensable role in sustaining life, development, and the environment. This includes surface and underground waters, in which the underground water includes thermal and mineral water. These types of water resources are a key identification of the economic growth and social prospect of a country [1–3]. Thermal and mineral waters (including spa waters) are known as valuable natural resources and are popular for medical therapy, such as the treatment of rheumatic diseases, skin diseases, arterial circulation disorders, muscular and skeletal damage, peripheral nerve damage, and other disorders [4]. Moreover, locations

with thermal and minerals water are suitable for tourism, recreation, and rehabilitation purposes as well [5]. Because of the popularity and importance of these types of water, their quality must be strictly controlled, including the concentration of radioactivity. For underground water, the physicochemical conditions and geological formation of the area strongly affects the occurrence of radionuclides, which can affect human health by the ingestion of drinking water from wells utilized in water-supply systems [1, 6, 7]. Therefore, the radiological characterization of drinking water has been a topic of concern of several organizations worldwide.

There are several standards and regulations determining limitations on radionuclide concentrations in water. However, measuring individual radionuclide activities costs a lot of time and is always expensive [3]. For this reason, there is a necessity for a method that is not only low cost, rapid and simple, but also it has to be applicable for determining safe drinking water limits. Thus, gross alpha - beta analyses are widely used as the first step of the radiological characterization of drinking waters [7–11]. The gross alpha activity in water mostly comes from the terrestrial radionuclides of the natural decay series such as ²³⁸U, ²³⁴U, ²³²Th, ²²⁶Ra. Gross beta is contributed mainly by the β emitting radionuclides in the thorium and uranium series (such as ²²⁸Ra and ²¹⁰Pb) and ⁴⁰K [12]. According to World Health Organization (WHO), the consumption of water for human is 2 liters per day and the annual dose rate is maximum is 0.1 mSv.year⁻¹. The water is considered radiologically safe, when total alpha and

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beta activity concentration is no more than 0.5 Bq L^{-1} and 1 Bq L^{-1} , respectively [13].

In the world, there are many reports about the levels of gross alpha and beta in mineral and thermal water sources in recent years. For example, Jobbágy et al. studied the gross alpha and beta levels in thermal water collected from Balaton Upland, Hungary [8]. They discovered activity concentrations ranging from 26 to 1750 mBq/L and from 33 to 2020 mBq/L for gross alpha and beta, respectively [8]. Bonotto reported results of the gross alpha and beta measurements and the estimated radiation dose from thermal and non-thermal underground waters [14]. In his study, the gross alpha ranged from lower than the detection limit (1 mBq L^{-1}) to 428 mBq L^{-1} , while the mean of gross beta was 0.4 Bq L^{-1} [14]. In Vietnam, so far no studies on the measurement of the radioactivity in thermal or mineral water sources (used for bathing, drinking and as mineral water) were presented before, with the exception of a limited scope study by Van Duong et al. [15], which contains information on radium and uranium isotope content in the water of the Kim Boi spa in Hoa Binh province. Investigation of the waters' characteristics, both chemical and radiological is an important task to assure public health and safety and provides baseline information for the local and regional governments in the area. Therefore, the main object of this study

is to determine the gross alpha and beta activity in eight spa and mineral water sources in northern Vietnam.

Experimental

Sampling and sample preparation

Water samples were collected at 8 spa and mineral water sources in northern Vietnam, including Quang Hanh, Quang Ninh province (QH-QN); Tien Lang, Hai Phong province (TL-HP); Kim Boi, Hoa Binh province (KB-HB); Thanh Thuy, Phu Tho province (TT-PT); Mi Lam, Tuyen Quang province (ML-TQ); Kenh Ga, Ninh Binh province (KG-NB); Ban Moong, Son La province (BM-SL); and It Ong, Son La province (IO-SL) spa and mineral water sources. The location of the sampling sites is shown in Fig. 1. Most of the aquifers in the sampling locations are hosted in limestone formations aged from Triassic to Devonian. The oldest geologic formation is located in Mi Lam – Tuyen Quang province and the youngest one is at Kenh Ga - Ninh Binh province. These waters are frequently consumed as drinking water by the local population (used as tap water and for cooking without treatment, with an estimated daily water consumption of 2 L, they are also used for bathing and spa

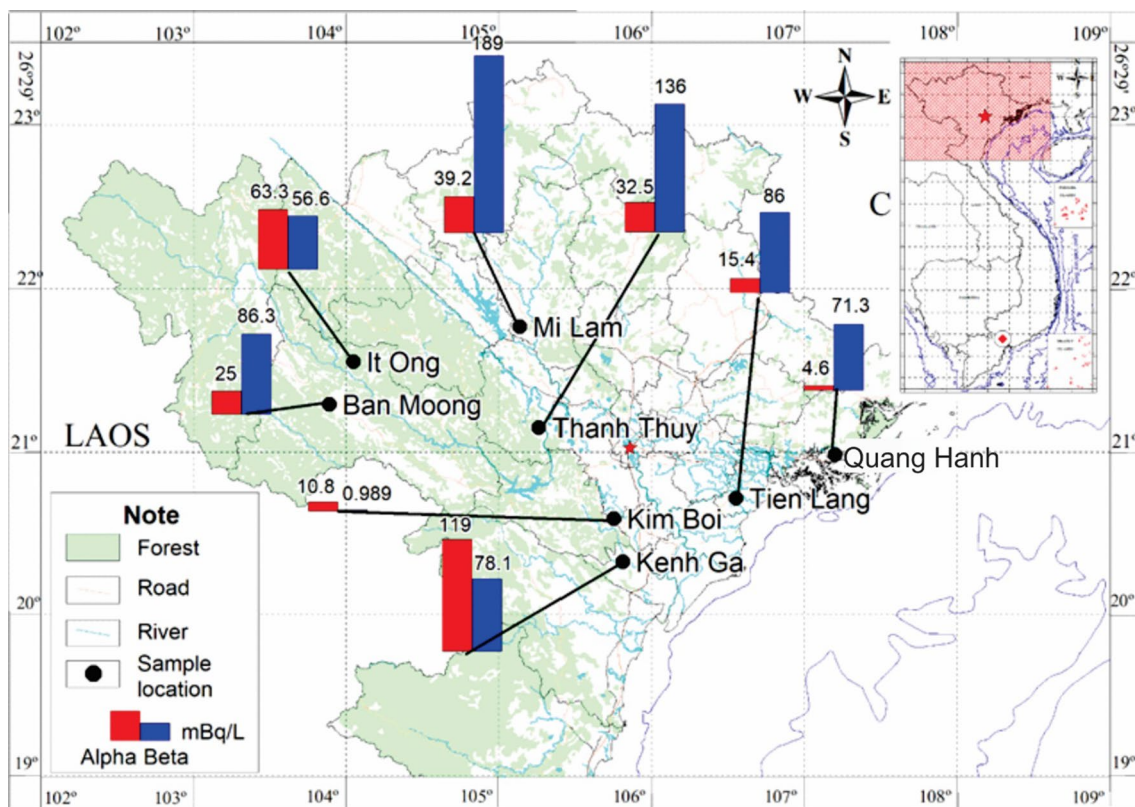


Fig. 1 Sampling locations

activities) and are used for therapy as mineral water. The exception is KG-NB, which is only consumed occasionally for stomach ailments. QH-QN, KB-HB, TL-HP, TT-PT are bottled and sold as mineral water as well.

The TDS as well as the temperature, EC (electrical conductivity) was measured immediately in situ after collecting the sample by a Hanna HI98194/40 multiparameter instrument, while pH was measured by a HI98190 pH meter.

Water samples of 10 liters each were taken using a polyethylene small barrel type sampling container directly from the source. Before using, the container was thoroughly cleaned. First, it was washed with washing liquid and rinsed out with tap water. After that, the container was soaked with 10% HCl acid for 10 h, then washed again with tap water until pH = 7. Finally, it was rinsed out three times using distilled water. The water samples were acidified to about pH = 2 with concentrated HCl acid to reduce the loss of radionuclides and the growth of microorganisms.

In the laboratory, from each sample three 1 L subsamples were taken into separate beakers, placed on a heating plate at about 90 °C, evaporated to below 50 mL, and diluted back to 50 mL at room temperature. After cooling, the concentrated water sample was transferred to a porcelain evaporating dish, then dried at 80 °C to constant weight. The sources are prepared by spreading a certain amount of residue evenly on the surface of the measuring tray for each measurement. To avoid the influence of sample's self-absorption on the measurement results, 0.05 mg/mm² and 0.1 mg/mm² were taken for gross alpha and gross beta measurement, respectively. These values were selected in order to achieve good counting efficiency and measurement stability. The tray radius of the measured samples is 25 mm and the sample mass of the residue is controlled at 196 mg ± 10%.

Gross alpha/beta activity concentration measurement

The samples were measured using a Canberra LB4100 low background A/B counter (Canberra Industries, USA). The system has an α and β background count < 0.10 cpm, and a β background count < 0.93 cpm. The detector is a proportional gas counter with a gas composition of 10% methane and 90% argon. The system is calibrated by ²⁴¹Am and ⁴⁰K standard surface sources (activity concentration of 14.7 Bq g⁻¹ and 16.1 Bq g⁻¹, respectively). The α-counting efficiency is 23%, and the β-counting efficiency is 41%.

The minimum detectable activity concentration of the system is determined by Eq. 1 [16, 17].

$$\text{MDC}(\text{Bq L}^{-1}) = \frac{L_d}{V * T * \epsilon * 60} \quad (1)$$

where $L_d = 2.71 + 4.65 \sqrt{(Bc * T)}$, V is the volume of the measured sample (the unit is liter); T is the measurement time (the unit is min); ε is the detection efficiency, and Bc is the background count rate (the unit is cpm). The minimum detectable activity concentration of the measurement system is calculated to be 1.8 mBq L⁻¹ and 1.6 mBq L⁻¹ for alpha and beta respectively. The measurement time of the sample, the standard sample and the background was equally 1000 min.

Measurement of chemical composition

Samples for the chemical composition measurement were collected with 100 ml plastic bottles filled to full without gaps and with tight lids. The chemical composition was analyzed in the laboratory after arrival using an ICP-OES PerkinElmer Optima 7300 DV spectrometer, calibrated with a multi-element standard solution of the Merck® company. "HCO₃⁻ was measured by classical acid-base titrimetry, while Cl⁻ and SO₄²⁻ were measured by Gallery Plus Discrete Analyzer - Thermo Fisher Scientific (spectrophotometry).

Result and discussion

The value of pH, total dissolved solids (TDS), gross alpha and gross beta activity, total cations and anions in the selected thermal water sources are presented in Table 1. The pH value in the thermal water sources ranged from 6.9 to 7.9, and the average value was 7.3. Most water sources are neutral, with the exception of the KB-HB and IO-SL samples, which are slightly alkaline. The temperature ranged from 39 to 60 °C, with an average value of 47 °C. The observed thermal water sources can be classified as warm and medium-hot mineral waters. The average value of TDS was 2650 mg L⁻¹ and it ranged from 319 to 9270 mg L⁻¹. Although, there is no reliable data on the possible health effects associated with the ingestion of TDS in drinking water, the presence of dissolved solids in water may affect its taste [18]. Specifically, the taste of drinking water is excellent, when the TDS level is less than 300 mg L⁻¹; good, between 300 and 600 mg L⁻¹; fair, between 600 and 900 mg L⁻¹; poor, between 900 and 1200 mg L⁻¹; and unacceptable if greater than 1200 mg L⁻¹. Therefore, the water at TL-HP, KB-HB and BM-SL are classified as good taste; the water at QH-QN is fair taste and the remaining are in the unacceptable category. The mean level of ions including HCO₃⁻, Cl⁻, SO₄²⁻, Na⁺, K⁺, Ca²⁺ and Mg²⁺ are 242, 725, 751, 460, 16, 332, and 116 mg L⁻¹, respectively, the range of these anions are 151–350, 4.21–4980, 5.6–1900, 9.3–2550, 0.1–46, 48.2–810, and 3.19–341 mg L⁻¹, respectively. The total anion and cation values in thermal water vary from 226 to 5670 mg L⁻¹ and from 65 to 3580 mg L⁻¹ with the mean value of 1720 mg

Table 1 The gross alpha, gross beta and chemical components concentration in thermal water mines

Sample (n=3)	pH	T ^o °C	TDS mg L ⁻¹	Gross alpha mBq L ⁻¹	Gross beta mBq L ⁻¹	HCO ₃ ⁻ mg L ⁻¹	Cl ⁻ mg L ⁻¹	SO ₄ ²⁻ mg L ⁻¹	Na ⁺ mg L ⁻¹	K ⁺ mg L ⁻¹	Ca ²⁺ mg L ⁻¹	Mg ²⁺ mg L ⁻¹	Total anion mg L ⁻¹	Total cation mg L ⁻¹
TL-HP	6.9	54	326	15.4	86.0	188	29.2	8.53	9.31	6.2	80	3.19	226	99
QH-QN	7.4	43	735	4.60	71.3	214	181	139	65.1	21.5	102	12.1	534	201
TT-PT	7.2	40	3150	32.5	136	151	312	1790	215	28.1	459	201	2250	903
ML-TQ	7.3	60	4060	39.2	189	262	259	1900	612	24.6	602	341	2420	1580
KG-NB	7.1	53	9270	119	78.1	350	4980	337	2550	46	810	174	5670	3580
KB-HB	7.9	44	354	10.8	0.99	239	11.3	9.25	9.4	1.29	73	10.5	260	94
BM-SL	7.1	39	319	25.0	86.3	239	4.21	5.6	11.5	0.1	48	5.6	249	65
IO-SL	7.6	41	3010	63.3	56.6	292	18	1820	206	1.01	485	182	2130	874
Average	7.3	47	2650	38.7	88.0	242	725	751	460	16	332	116	1720	920
Min	6.9	39	319	4.6	0.99	151	4.21	5.6	9.3	0.1	48	3.19	226	65
Max	7.9	60	9270	119.0	189	350	4980	1900	2550	46	810	341	5670	3580
Standard deviation	0.3	8	3070	37.3	55.4	62	1720	906	868	17	294	127	1870	1200

L⁻¹ and 920 mg L⁻¹, respectively. The highest total anion and cation values are found at KG-NB. The lowest total anion and cation values are observed in TL-HP and BM-SL, respectively. The chemical components decide the type of the thermal water sources. In this study, according to the Kurlov formula, the water source in QH-QN has sodium-calcium-sulfate-bicarbonate-chloride type water and KB-NB is sodium chloride type water. The water in these areas could be affected by salt intrusion from the sea. The remaining water sources have different types: TT-PT and IO-SL have calcium-magnesium sulfate type water, TL-HP, KB-HB, and BM-SL have calcium bicarbonate type water, and the water at ML-TQ is of the calcium sulfate type.

The mean activity concentrations of gross alpha and beta in the eight thermal water sources are 38.7 mBq L⁻¹ and 88.0 mBq L⁻¹, respectively. The min and max of gross alpha and gross beta activity concentrations are 4.6 and 119.0 mBq L⁻¹ and 0.99 and 189 mBq L⁻¹. The gross beta activity was approximately 3 times more than the amount of the gross alpha activity at most locations. However, the gross alpha levels in the water of 3 water sources, namely KG-NB, KB-HB and IO-SL, are higher than the gross beta level. The activity of gross alpha is much lower at QH-QN, which has sodium chloride type water, with the concentration of 4.6 mBq L⁻¹. The lowest gross beta activity concentration was found at KB-HB, 0.99 mBq L⁻¹. The KB-HB samples have sulfate - bicarbonate calcium type water, with medium mineral content. The highest gross alpha activity was found at KG-NB with 119 ± 17 mBq L⁻¹, which source has sodium chloride type water, and a high mineral content. The highest gross beta activity, 189 mBq L⁻¹, was observed in the ML-TQ water which is a sodium bicarbonate type water and has low mineral content. The activity concentration of gross alpha and gross beta are less than the screening level of 0.5 Bq L⁻¹ and 1 Bq L⁻¹ recommended by the World Health Organization [19], respectively. As mentioned in introduction, the underlying geological formation is one of the important factors that affect the radionuclide level in thermal water sources. In the current case, the geological characteristic at the thermal water sources are similar, and the underlying limestone aquifer has relatively low activity concentrations [10, 20, 21].

The gross alpha and gross beta concentration in this study meet the recommendations of WHO (less than 0.5 Bq L⁻¹ and 1 Bq L⁻¹ gross alpha and gross beta concentration) [19]. The gross alpha and gross beta activity concentrations measured in this study were compared with similar measurements in other places in the world (Table 2), such as drinking water [22], spring water [23], bottled water [24] and underground water [25]. As shown in Table 2, the average values of gross alpha and gross beta in this study were often lower than in other places in the world, except for Turkey [21, 22]. The gross alpha and beta activity concentrations in

Table 2 Comparison results between this study and some areas in the world (mBq L⁻¹)

Countries	Water samples	Gross alpha (mBq L ⁻¹)	Gross Beta (mBq L ⁻¹)	References
Vietnam (Northern)	Underground water	4.6–119 (37.3) ^a	0.99–189 (55.4)	This study
Iran (Guilan)	Well and spring	12–115 (52)	23–332 (110)	[22]
China (Haihe River Plain)	Underground water	17–362 (112)	18–779 (171)	[25]
Jordan	Underground water	180–9460 (1570)	360–7480 (1620)	[26]
Turkey	Bottled water	8–101 (21)	17–177 (59)	[24]
Nigeria (Kaseno state)	Surface and groundwater	24–665 (142)	7–1330 (285)	[27]
Turkey (Nevşehir province)	Underground water	13–182 (88)	81–779 (305)	[28]
Turkey (Trabzon)	Spring water	(8) ^b	(25)	[23]

^aRange: minimum – maximum (average)^bAverage

the water samples in this study are slightly lower than that in bottled water in Turkey and in spring water in Trabzon (Turkey). It should be noted that the bottled water has been processed before using, which leads to reduction in the gross alpha and beta activity concentration. For the spring water in Trabzon (Turkey), the relatively low gross alpha and beta can be attributed to the movement of water in the spring. Other places, such as Jordan and Turkey (Nevşehir province) have much higher gross alpha and gross beta levels. This is partially due to the underlying geological formations. In Jordan, the geological structure is heterogeneous, the level of gross alpha and beta is different in different areas, especially in Aqaba governorate. The high radionuclide level in groundwater in Jordan is attributed to the sandstone rock, which contains abundant thorium-enriched heavy mineral aggregations [26]. The high radionuclide level in groundwater in Turkey (Nevşehir province) is also attributed to the geological structure. This area is located in a very large plateau formed by the accumulation of ashes and lava of ancient

volcanoes in the Central Anatolia [24]. In contrast, the geological structure in this study area is limestone that has a lower radionuclide concentration. In addition, the observed radionuclide levels may be also affected by the precipitation rate. More precipitation will dilute the groundwater and reduce the concentration of the total gross alpha and gross beta as well as radioactivity level [25].

Table 3 presents the Pearson's correlation coefficients between gross alpha, beta, TDS, and total anion and cation concentrations. Based on Table 3, the gross alpha level is related to TDS, HCO₃⁻, Cl⁻, Na⁺ and Ca²⁺, while gross beta level shows correlation to Mg²⁺. The correlation coefficient between gross alpha and TDS is 0.94 that means TDS is strongly correlated to the gross alpha activity concentration. In other words, the gross alpha relates to the geological conditions that affects TDS. Moreover, the gross alpha is correlated to a number of anions and cations such as HCO₃⁻, Cl⁻, Na⁺ and Ca²⁺ that mean gross alpha activity is likely to be higher with water sources which have a higher concentration of these

Table 3 Pearson's correlation coefficient between gross alpha, gross beta and chemical composition

	pH	TDS	Gross alpha	Gross beta	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
pH	1	-0.21	-0.19	-0.53	0.15	-0.28	0.12	-0.26	-0.35	-0.13	-0.02
TDS		1	0.94**	0.28	0.69	0.89**	0.32	0.95**	0.82*	0.94**	0.64
Gross alpha			1	0.09	0.79*	0.87**	0.24	0.91**	0.61	0.86**	0.50
Gross beta				1	-0.17	-0.02	0.61	0.11	0.44	0.45	0.72*
HCO ₃ ⁻					1	0.69	-0.01	0.74*	0.28	0.59	0.29
Cl ⁻						1	-0.14	0.98**	0.77*	0.69	0.23
SO ₄ ²⁻							1	0.01	0.20	0.61	0.87**
Na ⁺								1	0.80*	0.79*	0.40
K ⁺									1	0.74*	0.51
Ca ²⁺										1	0.85**
Mg ²⁺											1

**Correlation is significant at the 0.01 level (2-tailed).

*Correlation is significant at the 0.05 level (2-tailed).

anions and cations. The gross beta level is correlated only to Mg^{2+} cations. As shown in Table 3, the correlation between gross alpha and beta activity concentrations in this study is weak, with a coefficient value of 0.09. The precipitation rate in North Vietnam is very high, with an annual rainfall of more than 1600 mm on average, and, especially in some mountainous areas, the annual rainfall can reach 2400–3200 mm (Vietnam National Transport Strategy Study-VITRANSS, 2000). As stated by Yi et al. [25], the high precipitation rate can cause weaken the correlation between gross alpha and beta due to the differences in its impact on the uranium and thorium decay chains relative to ^{40}K . The correlation coefficients observed during this study have some differences compared to the literature. In particular, multiple previous studies observed a strong relationship between the gross alpha and beta activity concentrations [24, 27, 28]. The gross alpha and beta activity concentrations also reportedly show an insignificant relationship with pH values and EC values of groundwater [26]. The correlation observed between gross alpha and gross beta in China was weak ($R = .43$) [25]. Yi and his colleagues observed a weak correlation between TDS and gross alpha and gross beta of the groundwater of $R^2 = 0.48$ and $R^2 = 0.22$, respectively [25]. The pH of the groundwater had no specific effect on the gross alpha and gross beta activity in Chinese underground waters [25].

Conclusions

The radioactivity concentrations in various mineral water sources distributed in North Vietnam have been extensively investigated in this study. The gross alpha and beta activity concentrations in the water samples in this study satisfy the WHO regulation for drinking water. The gross alpha has a strong correlation with TDS and some anions and cations (HCO_3^- , Cl^- , Na^+ and Ca^{2+}) while the gross beta has a strong correlation only with the cation Mg^{2+} . The gross alpha and beta concentrations have a weak correlation. The data obtained from this investigation can provide a baseline database for future research.

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