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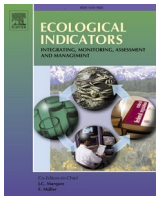

Chemical composition, radioactive and stable isotopes in several selected thermal waters in North Vietnam

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ABSTRACT

This work presents the chemical composition and concentration of radioactive and stable isotopes in water samples collected from nine well-known therapeutic centers located in north Vietnam. Excluding the Thanh Thuy – Phu Tho waters, which are hosted in Pleistocene sandstone with marble and gravel, the rest of studied waters are hosted in Paleozoic limestone formations. The thermal waters are extracted from 10 to 400 m below the surface, but the Keng Ga Ninh Binh intake is of an artesian nature. The temperature measured in the outlet waters ranges from 39 °C to 61 °C. The $\delta^{18}O$ and $\delta^{2}H$ (δD) vary from −9.13 to −5.09 ‰ and −63.9 to −34.1 ‰ respectively. All the thermal waters examined originate from infiltration water. This is also confirmed by the values of selected calculated hydrochemical indices. The hydrochemical types of the waters are HCO_3-Ca-Mg, HCO_3-Na, Cl-Na, SO_4-HCO_3-Ca-Mg, SO_4-Ca-Mg and SO_4-Ca. The mineralization of the concerned waters varies from 244 mgL^{-1} to more than 11,500 mgL^{-1}. The concentrations of uranium and radium in the waters are in the broad interval from about 4 $mBqL^{-1}$ to 680 $mBqL^{-1}$ for ^{238}U and from below 5 $mBqL^{-1}$ (limit of detection - LOD) to above 3400 $mBqL^{-1}$ for ^{226}Ra. There is no ^{228}Ra in most of the waters (below 10 $mBqL^{-1}$- LOD), excluding two waters in the Thanh Thuy-Phu Tho district, where ^{228}Ra is present with 18 and 27 $mBqL^{-1}$.

In all the waters studied, there is a very low concentration of polonium (^{210}Po) which varies from 0.5 $mBqL^{-1}$ to 8.2 $mBqL^{-1}$. High mineralization and high radium concentration should be associated with a long residence time in the given geological formation. The linear relationship between the depth of the host water formations and the water temperature suggests that the study waters are mostly heated from the geogradient of heat energy. But the water from the My Lam district is probably heated by intrusive magmatic fluid.

From a radiological point of view, the waters used for therapeutic and relaxation purposes are safe, however, there would be some problems if they were to be used for drinking purposes.

1. Introduction

There are more than 400 thermal water sources recorded in Vietnam with a temperature from 30 to over 100 °C. Thermal water is often used for medical treatment, sauna therapy and relaxation. Due to the high temperature, thermal water can interact strongly with its host geological formation and not only leach useful elements such as Ca, Mg, Fe, etc., but also the harmful heavy elements as Cu, Cd, Pb and radionuclides. These elements can induce a cancer of kidney and leukemia under prolonged exposure (Arfsten et al., 2007; Zhu et al., 2009; Hoffman et al., 2021). The assessment of water quality in terms of radioisotope and heavy element safety is one of the standard requirements (WHO, 2006, Lal et al., 2020). Knowledge of the concentration of radionuclides in groundwater, their behavior and their relation to the conditions prevailing in groundwater can bring new approaches for the study goals such as the origin, interaction processes and classification of thermal waters (Almeida et al., 2004; Focazio, 2001; Kasić et al., 2015; Sherif and Sturchio, 2018; Shin et al., 2016; Shivakumara et al., 2014).

During movement, a change occurs in the chemical composition and temperature of groundwater. The origin and behavior of key cations and anions in groundwater can improve our understanding of the geochemical behavior of groundwater and the interaction mechanism between water and the aquifer formation. Concentrations of cations and anions can be informative about geochemical processes within the

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aquifer and find groundwater pathways from the recharge to the discharge zone (Cook and Herczeg, 1999; Güler and Thyne, 2004; Huang et al., 2018; Jia et al., 2020; Rao et al., 2019). Many scientists around the world have concerned about the natural radionuclides in thermal water sources and reported wide ranges of radioisotope concentrations in different aquifer environments (Bonotto and de Oliveira, 2017; Condomines et al., 2012; Kovács-Bodor et al., 2019; Regenspurg et al., 2014; Scheiber et al., 2012; Tabar et al., 2013). The clear relationships observed between natural radionuclides and the chemical components in groundwater have shown that the presence of natural radionuclides is closely related to the geological and hydrogeological features of the aquifer (Almeida et al., 2004; Chau et al., 2016; Chau and Nowak, 2021; King et al., 1982; Sherif and Sturchio, 2018; Shin et al., 2016; Shivakumara et al., 2014; Stalder et al., 2012; Vespasiano et al., 2012). Uranium in groundwater is controlled by many factors e.g. high concentrations may be associated with formations containing U-rich minerals (Ayotte et al., 2007; Gascoyne et al., 1992; Langmuir, 1978; Keith et al., 2007; Smedley et al., 2006), or related to oxidizing conditions and CO_2-rich water (Jacobs and Smallinr, 2005; Jung et al., 2012; Smedley et al., 2006; Zhou and Gu, 2005). However, under reducing conditions, uranium can be removed from the water and precipitated into the rock formation (Jacobs and Smallinr, 2005; Wanty and Nordstrom, 1993). In some cases, the ratio of uranium isotope activity ($^{234}U/^{238}U$) is characteristic for the water resource and its analysis could be helpful in the determination of an aquifer forming a mixing zone (Osmond and Cowart, 1976; 2000; Osmond and Ivanovich, 1992; Suksi et al., 2001; 2006).

Radium is mobile in oxygen-poor, chloride-rich and highly mineralized groundwater (Kraemer and Reid, 1984; Zapecza and Szabo, 1986). The ratio of radium isotope activity ($^{226}Ra/^{228}Ra$) in groundwater is not only related to the relationship between uranium and thorium in the aquifer rocks, but can also provide information on residence time and the migration process (Asikainen, 1981a, b; Chau et al., 2012; Dickson, 1990; Sherif and Sturchio, 2018; Sturchio et al., 2001; Vengosh et al., 2009).

Radon has also been widely studied in mineral and thermal waters (Baskaran, 2016; Knutsson and Olofsson, 2002; Nandakumaran and Vinayachandran, 2020; Qadir et al., 2021; Vinson et al., 2009). The concentration of ^{222}Rn is a parameter that reflects the local geological and hydrological conditions. This gas can also be used in maintaining the migration paths and mixing processes of groundwater (Brutsaert et al., 1981; Erees et al., 2006; Moreno et al., 2018; Qadir et al., 2021; Seminsky and Seminsky, 2019; Sukanya et al., 2021).

For a long time, stable and radioactive isotopes have been used to study hydrological systems and have proven particularly useful for understanding features of groundwater systems such as origin and age, the influence of hydrogeological conditions, tectonic activity, climate and the environment of surrounding groundwater (Aggarwal et al., 2009; Zuber et al., 2004; 2008). Stable isotopes are considered one of the useful tools for studying the origin and recharge source of groundwater (Koeniger et al., 2016; Liu and Yamanaka, 2012; Liu et al., 2017; Praamsma et al., 2009). Furthermore, stable isotope studies allow groundwater contamination to be determined (Arumi et al., 2020; Barth, 1998) and water quality to be assessed (Kattan, 2018; Hassen et al., 2016; Zhou et al., 2016). Hydrochemical indices can complement radioisotope tests used to identify the origin of water. They are widely used in hydrogeochemical research. With their help, it is possible to determine the characteristic and origin of the tested water, as well as the processes responsible for the formation of its chemical composition. The formation of the chemical composition is a complicated process that takes place in different hydrochemical zones. Recognizing these zones contributes to, i.e. determining whether groundwater comes from the active supply zone with the inflow of fresh infiltration waters or from isolated geological structures. These processes are reflected in the values of hydrochemical indices, which are expressed as quantitative ratios of individual chemical compounds, that is, chlorides, bicarbonates, sulfates, bromides, iodides, sodium, calcium, and magnesium recalculated on milliequivalents (Lipiec et al., 2020; Abdelshafy et al., 2019; Han et al., 2009).

The relation between temperature and depth of the water aquifer can be used to evaluate the underground heat source (Chau and Nowak, 2021).

This work examines the nine known thermal water intakes in north Vietnam. The aims of the research are: (1) to assess the water quality and radiation safety; (2) to determine the characteristics, behavior and relationship between radioisotope activity and the physicochemical parameters of the thermal water and the aquifer's geological setting; (3) determine the origin of the waters and the waters' heat sources.

2. Material and methods

2.1. Regional geology and hydrogeology review

Vietnam is located in the Indochina Peninsula from 8° N to 22.5°N and has a tropical climate with an average air temperature from 26 °C to 32 °C and relative humidity from 74.1% to 78.9%. Therefore, thermal water in this country is regarded as a groundwater when the temperature at its output is higher than 30 °C (http://ceviwrpi.gov.vn/FileStorage/File/FileContent/Diachatthuyvan-thuatnguvadinhnghia.pdf). North Vietnam is divided into the North-East, North-West and Lowland Red River Delta geographical regions (Tri, 2016). The boundary of the North-East and North-West areas is the Chay River zone (Fig. 1). The North-East part is confined to the north by the boundary with China, to the south-east by the Bac Bo gulf and to the north-west by the Chay River (Fig. 1).

The following thermal water sources are found in the North-East region: Thanh Thuy – Phu Tho (MN-02, MN-03), My Lam – Tuyen Quang (MN-05) and Quang Hanh – Quang Ninh (MN-09). The North-West region borders with China to the North and with the Ma River and Laos to the West. The Kim Boi – Hoa Binh (MN-01), Ban Moong – Son La (MN-06) and It Ong – Son La (MN-07) sources are located within this region. The following thermal water sources are found in the Red River Delta Lowland: Kim Boi – Hoa Binh (MN-01), Keng Ga – Ninh Binh (MN-04) and Tien Lang – Hai Phong (MN-08).

Formations from the Paleozoic up to the Quaternary are present in

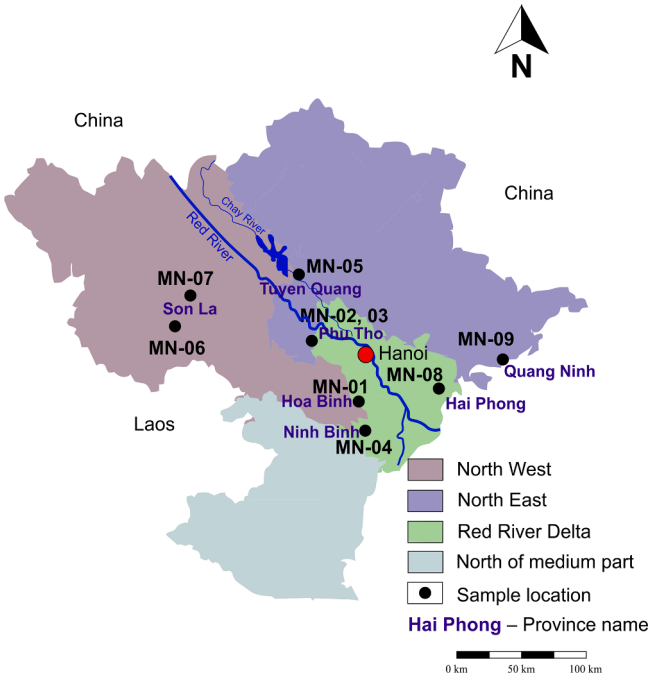

Fig. 1. The sampling locations in relation to the different geographical regions of North Vietnam.

the North-East region. The Paleozoic formations are made up of rocks such as: mudstone with mica schist, marble, limestone and dolomite with thin layers of fine-grained sandstone and shale which form the basement rocks. The Devonian, Carboniferous and Permian carbonate formations are dominant on most of the area of this region, especially in the east. The rocks occur as massif blocks and are strongly fractured. Mesozoic formations (Triassic, Jurassic and Cretaceous) are present as silica shales with thin layers of sandstone and as conglomerates. They lie unconformably on the Paleozoic formations. The Cenozoic terrestrial sediments are present as conglomerates, coarse-grained sandstone and clay with layers of brown coal and basically occur in the west of the zone close to the Red River delta. In this zone effusive and intrusive formations are also observed. The effusive rocks occur as thin rhyolite dykes close to the boundary with China. The intrusive rocks are represented by Paleozoic metamorphic granite-gneisses, biotite granites and syenite and occur along the Chay River zone. In the North-East zone, fractured Paleozoic and Mesozoic carbonate rocks and Quaternary sediments are the chief water host formations.

In the North-West, geological formations from the Archaic up to the Quaternary are present. Archaic gneiss, biotite schist and amphibolite are the main constituents of the Suoi Chieng formation which is more than 2000 m thick (McLean, 2002). Neoproterozoic rocks are

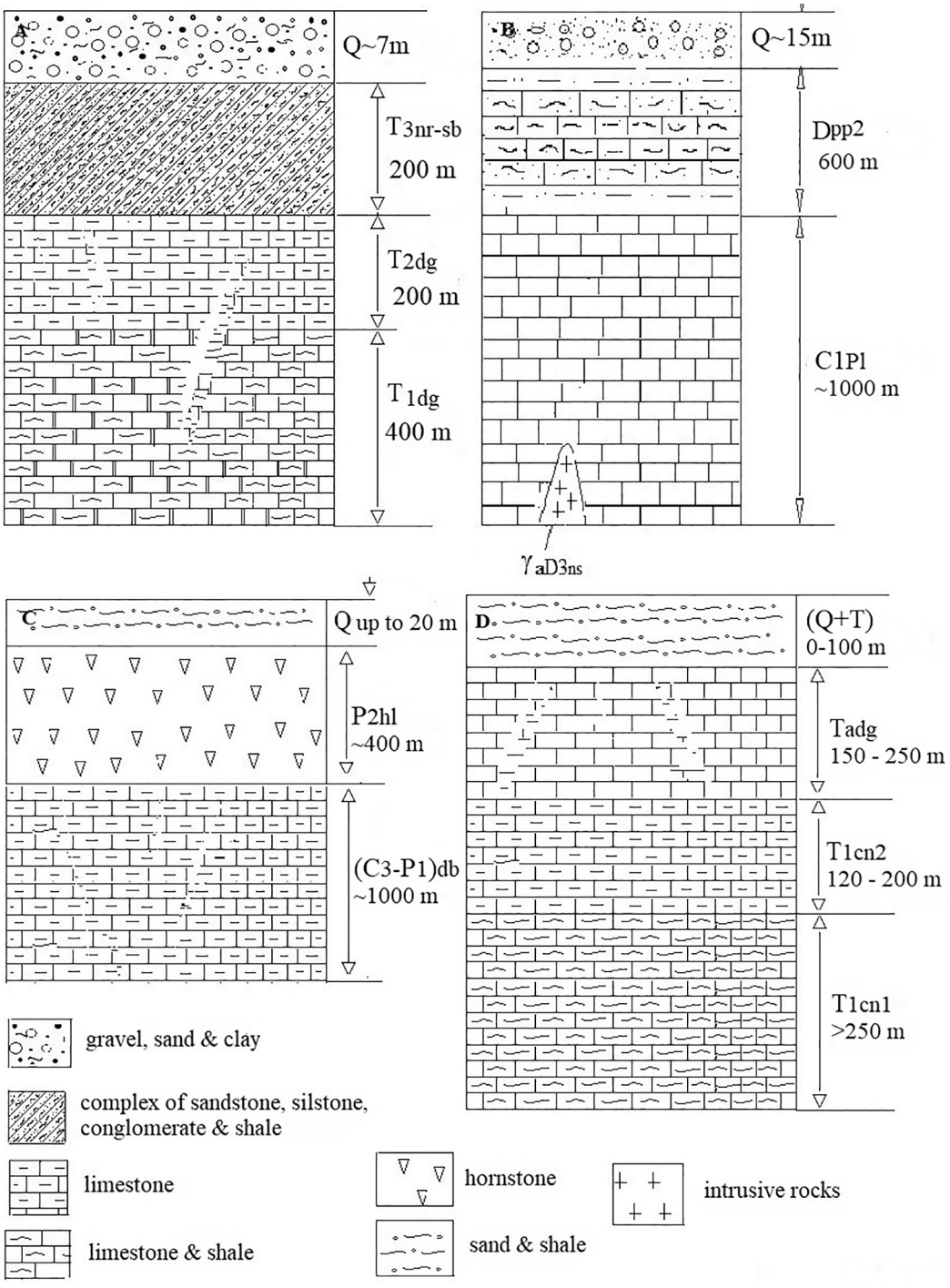

Fig. 2. Water host formations at the water intakes examined, A: MN-02, MN-03; B: MN-05; C: MN- 04, MN-08, MN-09; D: MN-01, MN-06, MN-07.

represented by metamorphic gneiss, two mica schists, amphibolite, garnets and graphite. The Paleozoic formations basically consist of mudstone with gravel, fine-grain sandstone and organic limestone. The Mesozoic formations are represented by two types of rock: effusive and sedimentary. The effusive rocks are basalts, andesite, and rhyolite. The tertiary carbonate sediments of the Lower and Middle Triassic belong to the Co Noi and Dong Giao formations. These formations are covered by Upper Triassic mudstone and fine-grain sandstone. The Quaternary sediments in this zone only occur on the river lowlands. In the North-West zone magma activities are found in the Neoproterozoic, Proterozoic, Paleozoic and Mesozoic epochs. These rocks are metamorphic amphibolite, granitoid, diorite, basalts and gabbro-diorites. They mainly occur in the Red River fault zone. The principal formations hosting waters are the surface deposits of Quaternary sand and the Dong Giao Triassic carbonate formation.

The Red River lowland is primarily composed of Cenozoic sediments. On the surface there are Quaternary alluvia, which lie on the Paleogene sediments occurring on most of the area. Outcrops of the older formations (from the Proterozoic up to the Jurassic) are only observed on the boundaries of the Red River Lowland. The Proterozoic formations are mainly represented by metamorphic gneiss, biotite schist and amphibolite nearly 2000 m thick. These rocks can be seen to the north of the Lowland. The Paleozoic formations consist of quartz schist, medium- and fine-grain sandstone, conglomerate, limestone with shale and lenses of coal. The above-mentioned rocks do not occur everywhere and their composition can vary from one place to another.

2.2. Formations hosting the thermal waters studied

The Thanh Thuy – Phu Tho thermal waters (MN-02, MN-03) are generally located in valleys surrounded by limestone mountains. The water host formation of these thermal waters is the Mid-Triassic Dong Giao fractured limestone formation. This formation occurs at different depths, from a few tens of meters to several hundred meters. The Dong Giao formation is covered by the Upper Triassic complex of conglomerates, sandstone, siltstone and shale with varying thicknesses. On the top of the formation there is Quaternary gravel, sand and a few meters of clay (Fig. 2A).

The thermal water in My Lam – Tuyen Quang (MN-05) occurs in fractured metamorphic crystalline carbonates of Devonian age (recrystallized limestone, marble with calcite and sericite mudstone). In this area Proterozoic metamorphic granite-biotite, amphibolite and gneiss are also present. On the top of the Devonian formations there is Quaternary sand, mud and gravel (Fig. 2B).

The thermal water at Quang Hanh – Quang Ninh (MN-09) is located near the Bac Bo Gulf (East Sea). The main thermal water aquifer is the strongly fractured Devonian-Carboniferous and karstic Deo But limestone formation. This limestone formation outcrops to the north and west of the North-East zone and forms mountain peaks with fractures and karsts reaching from a few tens of meters up to a few hundred meters in depth. In some places in this zone there are Quaternary gravels, sands and muds where sweet water occurs (Fig. 2C).

The thermal waters from Kim Boi – Hoa Binh (MN-01), Ban Moong (MN-06) and It Ong – Son La (MN-07) belong to the North-West zone and are hosted in the Dong Giao fractured limestone of Mid-Triassic age. In the Son La province the Dong Giao formation occurs below the Quaternary gravels, sands and muds with thicknesses from a few to several tens of meters. Below the Dong Giao formation there is the Downer Triassic Co Noi formation, which is built up of quartz schists, red and violet fine/medium grain sandstones together with thin carbonate rocks (Fig. 2D). In the Hoa Binh district the Mid-Triassic Dong Giao is situated in a similar manner to the Thanh Thuy – Phu Tho.

The Keng Ga – Ninh Binh (MN-04) and Tien Lang – Hai Phong (MN-08) thermal waters are located in the Red River Lowland. Many mountains formed of fractured limestone are also observed in the Ninh Binh province, the infiltration water migrating down and being heated by the geogradient. Then, under hydrostatic pressure, the hot water migrates up to the surface and creates an artesian water source.

Thermal water in the Tien Lang district, Hai Phong province (MN-08) is hosted in the fractured dolomite of the Deo But formation of Carboniferous age, which occurs at a depth of 400–800 m. The water host formation is covered by a hornstone formation with a thickness of nearly 400 m, and on the top of this there are Quaternary alluvia (Fig. 2C).

2.3. Samples collection and field measurements

The sampling bottles made of the polyethylene terephthalate (PET) were twice washed and rinsed with distilled water the day before sampling. In the field, the bottles were rinsed twice more with the thermal water collected. The water samples were placed into the previously prepared bottles and 5 ml of a two moles nitric acid was added to avoid precipitation. The temperature, electrical conductivity and pH of the water were measured directly by WTW/Cond 340*i*™. All the water samples were packed and sent to the AGH University of Science and Technology, Kraków, Poland, where they were analyzed. The locations of the sampling points were determined using a Garmin 60CSx GPS, with an accuracy of ±2–3 m and are shown in Fig. 1.

2.4. Measurement of chemical composition

Most of the elements in the water sample, such as Ca, Mg, Na, K, Mn, Fe, Sr, Zn, B, Ba, Ag, Cu, Ni, Co, Pb, Cd, Al, Cr, Mo, V, As, P and S, were determined using the inductively coupled plasma – optical emission spectrometry (ICP-OES) technique using an Optima 7300 DV spectrometer from Perkin Elmer™. The multi- and single element standard solutions of Sigma Aldrich® were used for preparation of the calibration curve. The PO_4^{3-}, SO_4^{2-} and SiO_2 concentrations were calculated based on the data on the total respective concentrations of P, S and Si determined from the analysis. The analytical procedure was performed according to the ISO 11885 standard. The Cl^- and HCO_3^- anions were measured using titration methods according to the ISO standards: ISO 9297 and EN ISO 9963-1 respectively. Certified Reference Materials – TMDA-64.3 and HAMIL 20.2 obtained from Environment and Climate Change Canada were used for quality control. The low limit of quantification (LOQ) of the methods for the major ions noted above is a tenth of a ppm and for the trace elements is several or several dozen ppb. The precision of the analytical methods at the LOQ level do not exceed 10% and the recovery is in the range of 80–120%.

2.5. Measurement of natural radionuclides

Uranium isotopes were co-precipitated from the ½ liter water sample together with manganese oxide MnO_2 after adding ammonium, $MnCl_2$ and $KMnO_4$. After cleaning up, the precipitate obtained was divided into two parts and both were weighed by analytical weight. One part was dedicated for uranium analysis and the second for polonium analysis. The part of the precipitate for uranium measurements was dissolved in 9 M HCl and then analyzed by ion exchange chromatography (IEC) using Dowex Cl^- resin. The uranium in the eluate was again precipitated after adding Mohr salt, $NdCl_3$ and HF. The obtained precipitate was placed onto a plastic membrane filter with 0.1 μm porosity and measured with an alpha spectrometer with silicon semiconductor PIPS (Passivated Implanted Planar Silicon detector). The efficiency of the chemical procedure was checked by adding a known amount of solutions with traces of ^{232}U and ^{209}Po isotopes at the very beginning of the precipitation procedure. The chemical procedure mentioned is described in detail in the publication of Skwarzec et al. (2003).

After adding disodium EDTA, $BaCl_2$, $Pb(NO_3)_2$ and sulfuric acid, radium isotopes were co-precipitated from the ½ liter water sample together with barium sulfate. After cleaning with distilled water and centrifugation, the precipitate obtained was dissolved in a 0.25 M EDTA

alkaline solution, and radium was again precipitated by adding acetic acid. The new precipitate was again cleaned using distilled water, centrifuged and placed in a glass vial of 22 ml volume, mixed with a 12 ml scintillation gel cocktail (a Perkin Elmer™ product) and measured using a liquid scintillation counter with α/β discrimination. The ^{226}Ra content was determined by the count rates from the alpha channel, while the ^{228}Ra content was established using the count rates from the beta channel after reducing the beta contribution of the beta decay radionuclides of the ^{226}Ra group. This contribution can be calculated using the Bateman equation. The detailed radium chemical procedure and measurement is described in the publication of Chau et al. (2012), Hao et al., (2019).

The part of the precipitate used for polonium analysis was dissolved in a concentrated HCl solution. Polonium isotopes were deposited on a silver disk and measured using an alpha spectrometer with a silicon PIPS detector. The LOQs for uranium, radium and polonium isotopes are 0.5 mBqL^{-1}, 5 mBqL^{-1} and 0.5 mBqL^{-1} respectively. The relative standard deviation of the result for all the isotopes analyzed is below 3%.

2.6. Measurement of stable oxygen and hydrogen isotopes

The hydrogen and oxygen isotopes of a water sample were measured using an L2140-*i* spectrometer (Picarro, Santa Clara, CA, USA) coupled with an autosampler (ALS-G, Applied Instruments, Germany) and a vaporizer unit (A0211, Picarro, Santa Clara, CA, USA). The autosampler collects a predefined volume of the water sample to be analyzed (1.8 µl) and transfers it through septa to the vaporizer unit, where it evaporates. The water vapor is mixed with high-purity nitrogen and transported by a diaphragm vacuum pump to the optical cavity of the optical spectrometer L2140-*i*, where its isotopic composition is analyzed by measuring laser absorption irradiating the water vapor. The calibration and reduction of the memory effect that remains in the spectrometer is performed using de-ionized water and the standard solutions USGS 47 and USGS 48. The relative standard deviation for both δ^{18}O and δ^{2}H (δD) is below 3%. The method cited is described in detail by Pierchala et al. (2019).

3. Results and discussion

3.1. Hydrogeochemical characteristic of the thermal water in North Vietnam

The results of chemical composition analysis with measurements of the temperature, pH, electrical conductivity and hydrochemical types of the study waters are summarized in Table 1.

The temperature of the study water varies from 39 to 61 °C and the depth of the water host formation is from 10 to 400 m. The mineralization of the water lies in the broad range from near 300 mgL^{-1} to more than 11,500 mgL^{-1}. The waters belong to different hydrochemical types: bicarbonate–calcium-magnesium (HCO_3-Ca-Mg), sulfate-calcium-magnesium (SO_4-Ca-Mg), sulfate-calcium (SO_4-Ca), sulfate-bicarbonate-calcium-magnesium (SO_4-HCO_3-Ca-Mg), and chloride-sodium (Cl-Na) with pH values from 6.9 to 8.6 (Fig. 3).

3.2. Radionuclides and stable isotopes concentrations

The concentrations of the radionuclides and delta deuterium (δD) and oxygen (δ^{18}O) measured are presented in Table 2.

The concentration of ^{238}U ranges from 3.66 to 680 mBqL^{-1}; the activity concentration of ^{234}U in water is often higher than that of ^{238}U and varies from 9 to 957 mBqL^{-1}. The elevation of ^{234}U activity upon that of ^{238}U in natural water is well known from the nuclear recoil (Osmond and Ivanovich, 1992). The uranium concentration in the water MN-04 and MN-09 is higher than the maximum permissible value (180 mBqL^{-1}) for drinking water (UNSCEAR, 2000). The concentrations of uranium in several thermal waters in north Vietnam: Keng Ga (MN-04), It Ong (MN-07) and Quang Hanh (MN-09) are higher than the uranium concentration of the other thermal waters in the world (Table 3). The uranium elevation in these waters can be related with the rich uranium water hosting formation and redox condition prevailing in it (Osmond, 1980). The ^{226}Ra varies from below 5 mBqL^{-1} as a limit of detection (LOD) up to 3430 mBqL^{-1} while ^{228}Ra is absent in the study waters apart from waters MN-02, MN-03 from Thanh Thuy - Phu Tho, where there is 27 and 18.2 mBqL^{-1} respectively. Both the low concentration of ^{228}Ra and the high activity ratio of radium isotopes (^{226}Ra/^{228}Ra) in waters probably result from: (i) the very low thorium content (^{232}Th) in the limestone formations of the aquifer and (ii) stronger interaction of the high-temperature water with rock minerals in the aquifer (Chau et al., 2012). Both the uranium and the radium concentrations are very high in thermal waters Keng Ga (MN-04), It Ong (MN-07) and Quang Hanh (MN-09) suggest the very long retention of these waters under ground. The ^{226}Ra concentrations in the studied waters are within the range of that in the thermal waters in the world (Table 3), while ^{228}Ra concentration in the concerned waters is really lower than that in the thermal waters in the world (Table 3) suggesting the most water bearing formations in the north Vietnam is limestone. The amount of polonium is very low in the water analyzed and ranged from 0.56 to 8.26 mBqL^{-1}. Such low polonium contents were also observed in Polish mineral water (Skwarzec et al., 2003).

3.3. Identification of water origin using radionuclides, stable isotopes and hydrochemical indices

The δD vs δ^{18}O relation is shown in Fig. 4. All the points lie very close to the Crag line, suggesting that the waters studied originated from infiltration (Zuber et al., 2008). Due to their locations in the lowland and close to the sea, the δ^{18}O and δD concentrations of MN-04 (Kenh Ga – Ninh Binh), MN-08 (Tien Lang – Hai Phong) and MN-09 (Quang Hanh – Quang Ninh) are higher than in the sources: MN-02, MN-03 (Phu Tho), MN-05 (Tuyen Quang), MN-06 and MN-07 (Son La), which occur in mountain regions nearly 200 km distant from the Sea (Fig. 1).

The origin of the thermal waters studied is also confirmed by the hydrochemical indices which are helpful in identification of their origin and of the processes responsible for the formation of their chemical composition (Lipiec et al., 2020; Jasnos, 2021; Rusiniak et al., 2021). The concentrations of individual chemical compounds determined in the tested waters were further recalculated on milliequivalent units taking into account the valency and molar mass of each element or/and ion. The results of the milliequivalent ratios of the selected elements or ions are the hydrochemical indices presented in Table 4.

As seen in the results, the high $rHCO_3^-/rCl^-$ ratios calculated for the MN-01, MN-02, MN-05, MN-06 and MN-08 thermal water intakes indicate the presence of infiltration water from the Miocene level in the active supply zone, where the groundwater remains in good hydraulic contact with infiltration waters (Lipiec et al., 2020; Rosenthal, 1988). Furthermore, the very high $rSO_4^{2-}x100/rCl^-$ ratio observed in four intakes (MN-01, MN-02, MN-03 and MN-07) could result from active water exchange with infiltrating waters (typical values from 10 to 500) or the aquifer could be supplied by groundwater containing dissolved gypsum (Han et al., 2009; Pazdro and Kozerski, 1990). For MN-08 and MN-09, the sulfate index ($rSO_4^{2-}x100/rCl^-$) is 10.9 and 10.1 respectively, additionally for MN-09 the rNa^+/rCl^- index is 0.86 which are very close to sea water or ocean water. These values indicate no impact of younger waters on their chemical composition. If the major concentration of sodium and chlorides in groundwater is derived from the silicates weathering the rNa^+/rCl^- value is greater than 1. On the other hand if evapotranspiration or halite dissolution processes were responsible for Na^+ and Cl^- concentrations, then Na/Cl molar ratio should be close to 1 (Jankowski and Acworth, 1997; Han et al., 2009). As the Hounslow (1995) showed, the $rNa^+/(rNa^+ + rCl^-)$ ratio presents the origin of sodium ions. In the aquifer containing the clay minerals such as montmorillonite, natural softening of the water (decreasing of the water

Table 1
Physical and chemical properties and principal ion composition [mgL^{-1}] of selected thermal waters from north Vietnam.

Water intake	Intake name	XY coordinates	Depth[a]	Temp. [°C]	TDS [mgL^{-1}]	γ_{25} [$mScm^{-1}$]	Eh [mV]	pH	K^+ [mgL^{-1}]	Na^+ [mgL^{-1}]	Ca^{2+} [mgL^{-1}]	Mg^{2+} [mgL^{-1}	HCO_3^- [mgL^{-1}]	Cl^- [mgL^{-1}]	SO_4^{2-} [mgL^{-1}]	Hydrochemical type
MN-01	Kim Boi-Hoa Binh	X = 20°35′30″ Y = 105°35′10′′	157 m	44	292	0.343	229	7.7	1.47	5.12	39.9	14.8	100	6.5	92.3	SO_4-HCO_3-Ca-Mg
MN-02	Thanh Thuy-Phu Tho 1	X = 21°09′12″ Y = 105°15′41′′	25 m	39	2430	2.56	202	7.6	9.18	50.4	410	163	171	11.3	1357	SO_4-Ca-Mg
MN-03	Thanh Thuy-Phu Tho 2	X = 21°09′10″ Y = 105°15′40′′	55 m	41	3310	3.58	182	7.5	6.94	208	620	106	120	312	1850	SO_4-Ca
MN-04	Kenh Ga-Ninh Binh	X = 20°19′30″ Y = 105°48′30′′	artesian	53	8480	14.8	192	6.9	39.9	2290	588	177	95.2	4750	273	Cl-Na
MN-05	My Lam-Tuyen Quang	X = 21°46′10″ Y = 105°08′20′′	90 m	61	370	0.326	153	8.2	1.84	66.5	6.65	0.21	159	14.3	13.5	HCO_3-Na
MN-06	Ban Moong-Son La	X = 21°17′40″ Y = 103°53′20′′	10 m	39	244	0.212	159	7.8	0.71	6.29	26.6	9.60	147	4.50	4.91	HCO_3-Ca-Mg
MN-07	It Ong-Son La	X = 21°33′25″ Y = 104°02′50′′	47 m	41	3080	3.55	184	7.6	6.47	203	597	96.5	115	303	1700	SO_4-Ca
MN-08	Tien Lang-Hai Phong	X = 20°43′00″ Y = 106°33′00′′	400 m	55	474	0.497	175	8.5	4.37	104	9.22	2.75	246	30.9	4.54	HCO_3-Na
MN-09	Quang Hanh-Quang Ninh	X = 20°59′10″ Y = 107°11′50′′	210 m	43	11,520	18.8	234	7.7	110	3457	473	266	161	6168	842	Cl-Na

[a]data obtained from the owners of the water centers.

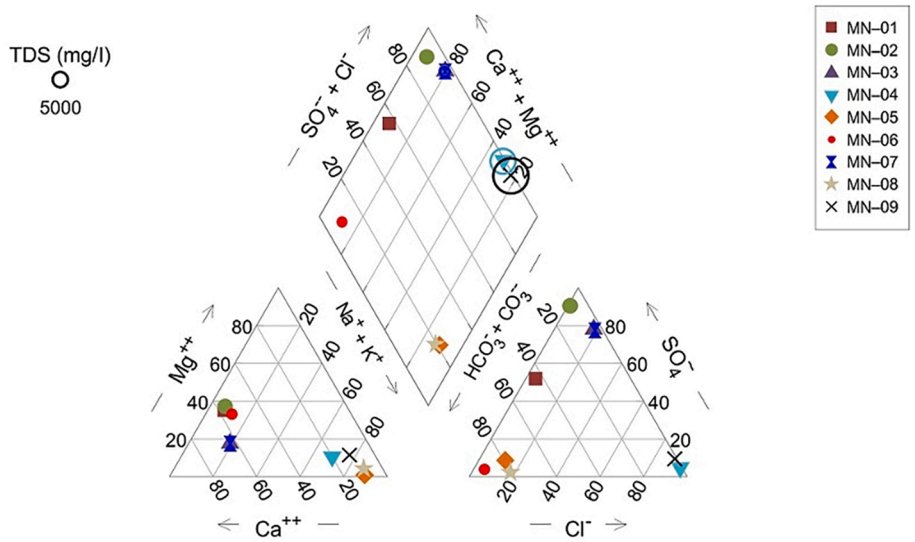

Fig. 3. Piper diagram presenting the chemical composition of the thermal waters studied.

Table 2
Measured concentrations of uranium, radium and radiolead as well as oxygen and hydrogen isotopes.

Sample	U-238 [mBqL^{-1}]	U-234 [mBqL^{-1}]	Ra-226 [mBqL^{-1}]	Ra-228 [mBqL^{-1}]	Po-210 [mBqL^{-1}]	$\delta^{18}O$ [‰]	δD [‰]
MN-01	3.66	18.6	16.5	≤ 10	1.01 ± 0.05		
MN-02	27 ± 4	112 ± 7	27 ± 4	27 ± 4	2.21 ± 0.19	−9.13 ± 0.25	−63.9 ± 1.9
MN-03	97 ± 1	9 ± 1	47,4 ± 5,0	18,2 ± 2,2	1.19 ± 0.09	−8.54 ± 0.21	−59.6 ± 1.5
MN-04	217 ± 12	280 ± 14	3430 ± 340	≤10	8.26 ± 0.66	−6,32 ± 0.16	−45.8 ± 1.1
MN-05	14 ± 3	26 ± 4	≤5	≤10	1.39 ± 0.12	−8.73 ± 0.22	−60.9 ± 1.6
MN-06	52 ± 6	90 ± 8	≤5	≤10	3.73 ± 0.22	−8.43 ± 0.21	−60.2 ± 1.6
MN-07	175 ± 14	367 ± 22	520 ± 50	≤10	0.56 ± 0.02	−8.56 ± 0.21	−59.9 ± 1.6
MN-08	11 ± 3	12 ± 3	14 ± 2	≤10	1.79 ± 0.18	−7.10 ± 0.20	−50.7 ± 1.5
MN-09	680 ± 33	857 ± 39	272 ± 27	≤10	2.83 ± 0.26	−5.09 ± 0.13	−34.1 ± 1.1

hardness) may occur. This process takes place when the calcium and magnesium ions are removed from water (adsorption onto the clay minerals) with the simultaneous release of sodium ions. Values of this index that are greater than 0.5 suggest that the Na ions originate from the ion exchange process with clay minerals found in marls or Neogene clays. On the other hand, values slightly higher than 0.5 suggest that the sodium in the water is not a product of halite dissolution, but its source may be an ion exchange between groundwater components and plagioclases (Hounslow, 1995). The source of Na^+ ions in the study waters is probably weathering of silicates or ion exchange with clay minerals that are present in the Quaternary and Paleogene-Neogene formations overlying the thermal water host rocks. For intakes MN-04 and MN-09 the value of rNa^+/rCl^- index is lower than 1. This can indicate the reverse ion exchange process, when the sodium ions are adsorbed onto the clay minerals and the calcium and sometimes magnesium ions are released to water. This process needs sodium-reach water which concentration is higher than clay-exchangeable equilibrium. This can be especially observed during the seawater intrusion (Hounslow, 1995). Contamination with seawater is also observed when the $rCl^-/(rHCO_3^- + rCO_3^{2-})$ is greater than 0.5 (Abdalla, 2016). The degree of groundwater contamination with seawater was divided into 6 water classes by Todd and Mays (2004) based on $rCl^-/(rHCO_3^- + rCO_3^{2-})$ index and who distinguished: good water (<0.5), slightly contaminated (0.5–1.3), moderately contaminated (1.3–2.8), injuriously contaminated (2.8–6.6), highly contaminated (6.6–15.5) and severely contaminated (>15.5). Among the thermal waters examined in this work, water derived from MN-1, MN-02, MN-05, MN-06 and MN-08 can be classified as good quality water due to the $rCl^-/(rHCO_3^- + rCO_3^{2-})$ value lower than 0.5 with no seawater intrusion. The MN-03 and MN-07 thermal waters can be considered as groundwaters mixed with seawater belonging to the injuriously contaminated class with the hydrochemical index value 4.47 and 4.53 respectively. For two intakes such as MN-04 and MN-09 the contamination with seawater is sever, due to the calculated molar ratio of $rCl^-/(rHCO_3^- + rCO_3^{2-})$ which is 85.86 and 65.92 respectively. For the MN-04 and MN-09 intakes, the thermal water origin can have two scenarios based on the hydrochemical indices calculated. The value of the rNa^+/rCl^- <1 calculated for MN-04 and MN-09 thermal waters indicates that the water exchange is limited, the reservoir is isolated and fresh water inflow is restricted which is characteristic of relict waters. It is also reflected in the $rHCO_3^-/rCl^-$ index which has a very low value. On the other hand $rCl^-/(rHCO_3^- + rCO_3^{2-})$ suggest the intrusion of seawater due to its very high value. In the light of the results of the stable hydrogen and oxygen isotopes analysis, the most probable scenario is the seawater intrusion due to the higher concentration of $\delta^{18}O$ and δD than in other intakes. A similar situation is observed in the MN-09 thermal water intake.

Figure 5 presents the relation between the depth of the water host formation and measured water temperature. Excluding water from My Lam – Tuyen Quang (MN-05), the temperature of the thermal waters studied and the depths of their host formations are linearly related at 0.038 ± 0.005 °C per meter. This fact suggests that the waters are heated by the geogradient with ~ 3.8 ± 0.5 °C per 100 m. In the case of My Lam – Tuyen Quang, the heat source is not geogradient but the magma fluid (Hiep et al., 2016).

An explanation of the hypotheses mentioned is based on the conceptual model describing the hydrogeological structure at My Lam – Tuyen Quang (Fig. 6). In the region near the surface there is a rock formation with very low water permeability (loess loam, shales); below

Table 3
Statistical values of TDS (total dissolved solids), temperature, ^{238}U, ^{226}Ra and ^{228}Ra activity concentrations of some selected thermal waters in the world.

Region	Number of samples	Statistic parameter	Temp. [°C]	TDS [mgL^{-1}]	^{238}U [mBqL^{-1}]	^{234}U [mBqL^{-1}]	^{226}Ra [mBqL^{-1}]	^{228}Ra [mBqL^{-1}]
Croatia	12	min.	22	n.a.	n.a.	n.a.	87	n.a.
(Marović et al., 1996)		max	96				6200	
		average	46				1322	
		median	40				283.5	
		st. dev.	23.2				385.8	
France	6	min.	16	3608	5	n.a.	588	260
(Rihs and Condomines, 2002)		max	41	5608	27		2287	1590
		average	30	4137	15		1158	854
		median	24	4137	16		947	794
		st. dev.	9.4	746.2	8.4		627.8	452.5
Poland (Nowak et al. 2012)	18	min.	22	201	≤0.5	≤0.5	19	23
		max	86	117,000	328	313	65,000	13,700
		average	47.9	14,405	29	32	7404	1806
		median	34	1385	3.1	7.6	437	79
		st. dev.	27	34,766	83.4	79.2	19,684	4382
Spain	19	min.	15	289	n.a.	n.a.	15	n.a.
(Duenas et al., 1998)		max	52	14,790			1367	
		average	28	3702			263	
		median	21	2070			157	
		st. dev.	12.4	4432.8			328.5	
Tunisia	24	min.	21	200	1.2	1.3	2	2
(Labidi et al., 2002)		max	75	24,600	69.1	153.4	1630	1032
		average	46	6604	9.6	18.3	507	177
		median	45	2840	4.3	7.0	358	113
		st. dev.	15.9	7606.3	15.5	33.0	489	217.6
Turkey	36	min.	29	n.a.	n.a.	n.a.	120	n.a.
(Belin et al., 2002)		max	90				700	
		average	51				337	
		median	47				315	
		st. dev	13.3				156.7	
Vietnam	9	min.	39	244	3.66	9	5	10
(this work)		max	61	11,520	680	857	3430	27
		average	43	2430	52	90	27	10
		median	46.2	3356	141.9	196.8	481.9	12.8
		st. dev	8.0	4033.2	215.7	278.5	1119.3	6.0

n.a. – data non available.

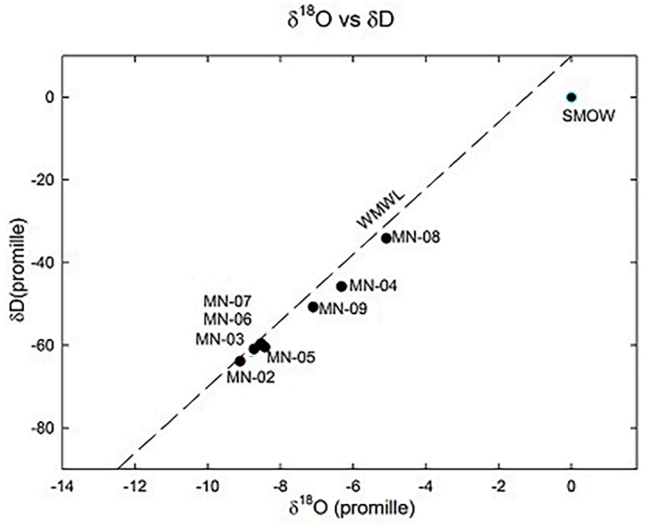

Fig. 4. Relation $\delta^{18}O$ and δD, WMWL – world meteoric water line, SMOW – standard mean ocean water.

this layer a conductive formation (fractured sandstone) occurs. Both formations are surrounded by the crystalline basement formation with fractures at the boundaries with the mentioned layers and a magma fluid underneath. When the infiltration water migrates down along the fracture paths, the water reaches the conductive layer and receives the heat energy from the magma fluid. As a result of static pressure, the water migrates up along the fault to the surface.

Excluding the water from It Ong – Son La (MN-07), there is a positive linear relation between ^{238}U and ^{226}Ra (Fig. 7). Such a positive dependence of radium on uranium in groundwater is rarely observed (Asikainen and Kahlos, 1979; Hakam et al., 2001), because in groundwater reducing conditions are often dominant where uranium is immobile, while radium is significantly lighter and controlled. Therefore in groundwaters the negative proportion between radium and uranium is observed (Hakam et al., 2001). In surface or soil waters, the concentration of uranium is often higher than that of radium. In water occurring in deep formation, the radium concentration is higher than that of uranium. This is connected with the fact that the cumulation of radium in groundwater is more and more due to leachability of this element from the rock minerals, whereas in underground formation there are often reducing conditions due to limitation with atmosphere, uranium becomes immobile and tends to precipitate from the water onto formation rocks.

Calcium is a principal ion in groundwaters, while radium belongs to the calcium group, therefore there is often a positive correlation between water mineralization and radium concentration (Fig. 8).

In a similar manner to the relationship between Ra and U, the MN-07 water also lies far from the relation line of TDS vs Ra. The phenomenon suggests that the formation hosting It Ong – Son La water (MN-07) may be rich in uranium, such as limestone with shale or organic materials. The Fig. 9 shows a positive correlation between the uranium concentrations and the water mineralization (TDS).

Due to differences in geochemical properties between uranium and the main components and generally groundwater reduction conditions (Eh < 0), the correlation between TDS and uranium is often negative (Asikainen, 1981b; El-Sharkawy, 2018). In our case, there is a positive

Table 4
Values of the hydrochemical indices explaining the origin of the thermal waters studied.

Thermal water intake	Values of hydrochemical indices				
	$\frac{rHCO_3^-}{rCl^-}$	$\frac{rSO_4^{2-}x100}{rCl^-}$	$\frac{rNa^+}{rCl^-}$	$\frac{rNa^+}{rNa^+ + rCl^-}$	$\frac{rCl^-}{rHCO_3^- + rCO_3^{2-}}$
MN-01	8.94	1048.7	1.21	0.55	0.112
MN-02	8.79	8869.0	6.87	0.87	0.114
MN-03	0.22	437.9	1.03	0.51	4.47
MN-04	0.012	4.2	0.74	0.43	85.86
MN-05	6.46	69.7	7.17	0.88	0.155
MN-06	18.98	80.6	2.15	0.68	0.053
MN-07	0.22	414.4	1.03	0.51	4.53
MN-08	4.63	10.9	5.19	0.84	0.216
MN-09	0.015	10.1	0.86	0.46	65.92
Reference value for ocean water	**0.007**[1)]	**10.3**[2)*]	**0.87**[2),3)]	**0.5**[4)]	—

rX is a mass concentration of the ion X in the water expressed in the equivalent weight unit.
[1)] Winid and Lewkiewicz-Małysa, 2010.
[2)] Pazdro and Kozerski, 1990 *sea water.
[3)] Macioszczyk, 1987.
[4)] Hounslow, 1995.

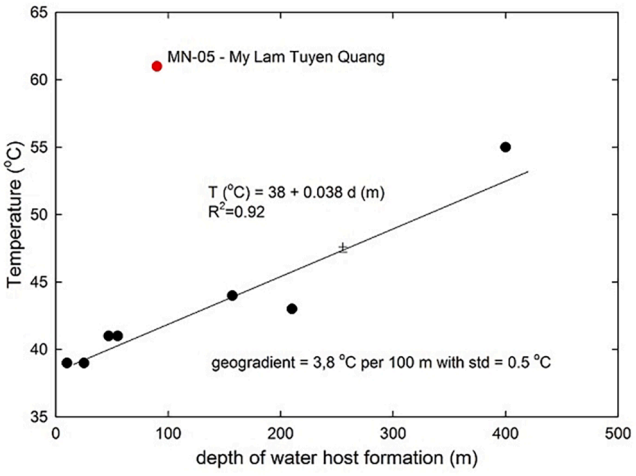

Fig. 5. Relation between the depth of the water host formation and the water temperature.

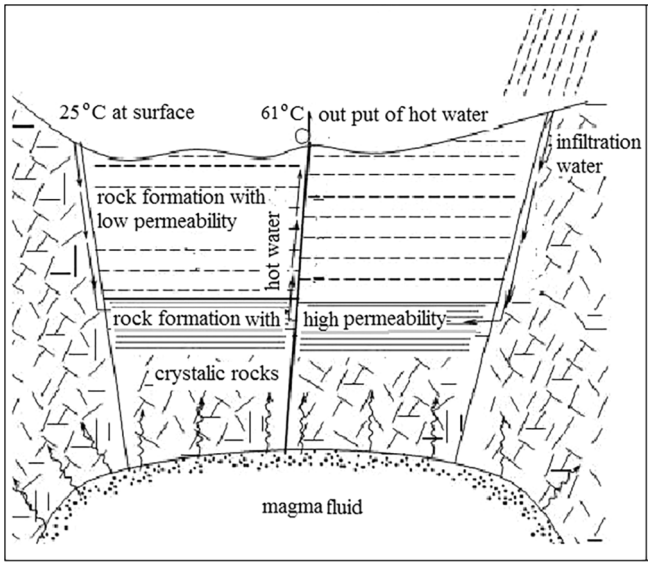

Fig. 6. Model of the hydrogeological structure in the My Lam Tuyen Quang district, modified from Hiep et al. (2016).

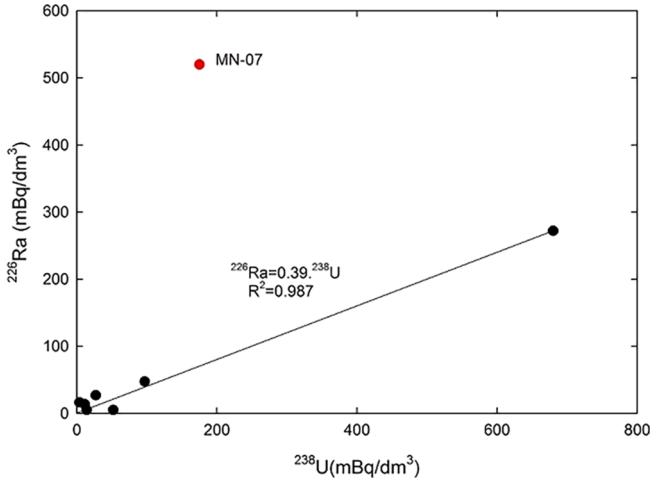

Fig. 7. Uranium-238 vs. Radium-226.

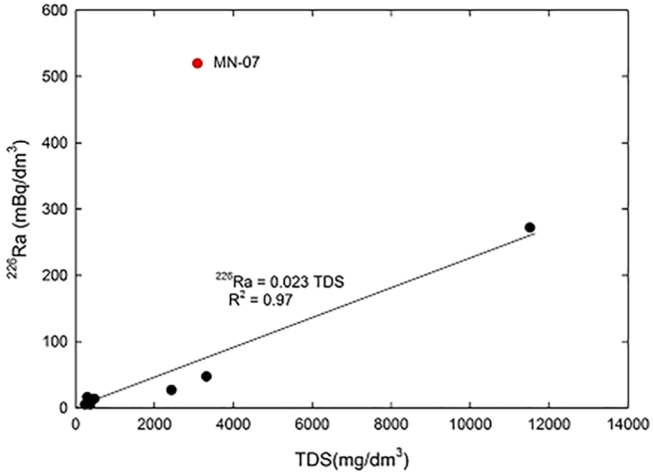

Fig. 8. Relationship between TDS and ^{226}Ra concentration.

proportion between TDS and uranium (Fig. 9) similarly to the relation between uranium and radium suggests the oxidising conditions prevailing at the studied thermal waters.

There is no correlation between mineralization and the depth of the aquifer (Fig. 10), suggesting that the hydrogeological conditions are

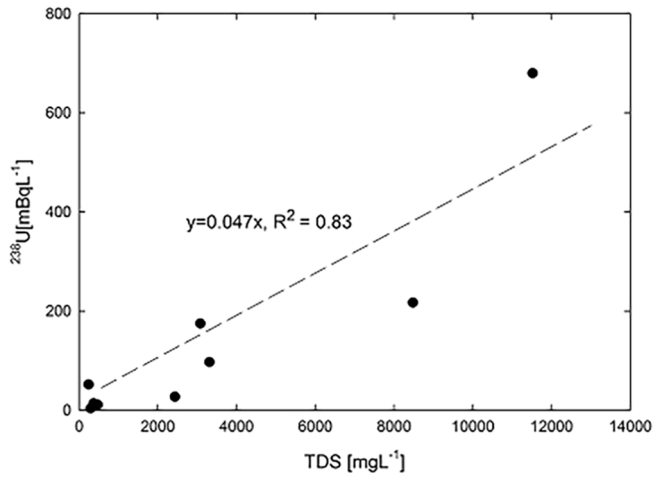

Fig. 9. Relationship between TDS and ^{238}U concentration.

different in the different aquifer formations.

The chemical composition of the thermal waters can be considered stable. The archives results on the example of the My Lam thermal water, which has been observed from the 1940s to the present (Table 5), are very similar to those presented in this work. Generally, the values of the physicochemical parameters (TDS, temperature, chemical compounds) seem to be stable. Although the measured values of each parameter differ from each other, the variations may be the result of the methods adopted for sampling and measurements. To prove the stability of the tested waters, the periodic long-term observations should be provided and the same sampling protocol and measurements methods should be used. This will avoid errors resulting from changes in sampler and laboratory measurements (Kmiecik, 2018)

3.4. Risk assessment connected with radionuclides and water quality

To appreciate the risk resulting from the use of the study waters, the annual effective dose received was estimated based on the assumption that every adult drinks two liters per day (WHO, 2004) and the formula is as follows:

$$D(g) = \sum_{i=1}^{n} (A_i e_i(g)) \bullet V \tag{1}$$

where: A_i is the activity concentration of the i-isotope in water, $e_i(g)$ is the effective dose received by a person in the g age group when the person absorbed one becquerel of the i-isotope. V is the volume of water

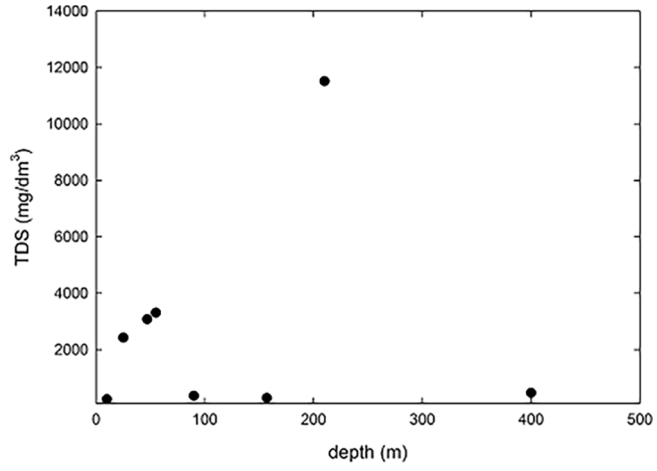

Fig. 10. Relation between aquifer depth and water mineralization (TDS).

consumed by the person. The values of $e_i(g)$ can be obtained from the International Basic Safety Standards (IAEA, 1996). The calculated doses for an adult who drinks two liters per day of the study waters under the assumptions adopted are summarized in Table 6.

Excluding waters from Kenh Ga – Ninh Binh (MN-04) and Ban Moong – Son La (MN-06), the effective doses received of the remaining waters are lower than the permissible dose (0.1 mSv/year). The doses from Kenh Ga and Ban Moong are higher than the maximum permissible level value, but the mineralization of these waters is higher than 3 gL^{-1}, so the waters are not classified as drinking waters and should not be used for human consumption. The contribution of ^{226}Ra in the total dose received is the largest and amounts to more than 70% (Fig. 11).

In the therapeutic case or relaxation, patients often live in the thermal water centers for a short time (for the patient - three weeks – 21 days and for the tourist – one week – 7 days per year). The use of water for the patient is in line with the doctor's recommendation (0.3 L per day), so the annual effective dose is at least 20 times lower than the calculated doses presented in Table 6. All doses for patients and tourists are far lower than 0.1 mSv/year, which means that the use of the thermal waters studied for therapeutic medicine and relaxation is totally safe.

The study of the natural radionuclides in the investigated thermal waters and assessment of the effective doses as a consequence of the absorption of the radionuclides in the water through consumption are in the scopes of point 3 and point 4 belonging to the 17 goals of sustainable development goals (SDG). The discussion concerning the underground energy sources that discharge thermal waters, as well as the energy of the thermal waters increasingly being used as a source of heating plates and electricity, is a topic of the points 6 and 7 of the 17 goals of the SDG.

In the case of using water for irrigation purposes, water is often evaluated by several quality parameters such as sodium absorption ratio (SAR), percent sodium (%Na) and residual sodium carbonate (RSC) parameters (Heleika et al., 2021). In this work regarding the major ions analysed, the SAR and %Na parameters were used which are defined as follows:

$$SAR = \frac{Na}{\sqrt{\frac{Ca+Mg}{2}}} \tag{2}$$

$$Na(\%) = \frac{Na + K}{Ca + Mg + Na + K} \times 100 \tag{3}$$

where: Na, K, Mg and Ca is the concentration of each element expressed in meqL^{-1} in the studied waters. The calculated values of the parameters concerned for every thermal water are summarized in Table 6. According to Heleika et al. (2021) and based on the SAR criterium waters from MN-04 (Keng Ga – Ninh Binh) and MN-09 (Quang Hanh – Quang Ninh) are bad quality (SAR greater than 18 and SAR greater than 26), and the rest of the waters can be classified as excellent waters (SAR $<$ 10). For MN-04 SAR is 21.9 resulting in classification to the doubtful water, whereas for MN-09 sodium absorption ratio is higher than 32 which makes this water unsuitable for irrigation purposes. Based on the sodium percent (%Na) criterium, five waters: MN-01, MN-02, MN-03, MN-06 and MN-07 are classified to the excellent category (%Na $<$ 20), MN-04 to doubtful (%Na from 60 to 80), and the rest thermal waters such as MN-05, MN-08 and MN-09 to the unsuitable group with %Na equal 89.7, 87.4 and 77.6 respectively.

4. Conclusions

Vietnam is a country located on the Indochina Peninsula and limited from 8°N to 22.5°N with tropical climate. In this country three fourth area is the mountain region and the average atmospheric precipitation is around 1400 mm per year. Additionally in the North Vietnam fissured limestone formations are prevailing, therefore this country is very rich in karst caves. Such conditions are enable for ground- and thermal water cumulation. Since the beginning of the 20th century, Vietnamese people

Table 5
Archival and current data from analyses of the temperature and chemical composition of the My Lam thermal water (Hiep et al., 2016).

Parameter measured	Autret (France) (1941)[a]	Vietnam Oil&Gas Department (1981)	Hanoi Univ. Pharmation (1984)	Czech Geol. Department (1988)	Hanoi Univ. Min. & Geol. (1999)	Vietnam Depart. Geol. & Mineral Resource (2012)	This work (2021)
Temp. [°C]	58.5	63	63	64	67	65.5	61
TDS [mgL^{-1}]	259	343	300	300	175	290	370
Na^+ [mgL^{-1}]	15	60	117	62.8	61.1	61.8	66.5
K^+ [mgL^{-1}]	3.70	4.10	3.83	2.40	3.60	2.30	1.84
Li^+ [mgL^{-1}]	n.a.[b]	n.a.	n.a.	0.08	n.a.	n.a.	0.041
Ca^{2+}[mgL^{-1}]	6.90	7.55	6.32	9.03	4.00	2.40	6.65
Mg^{2+}[mgL^{-1}]	3.12	6.41	2.50	1.70	0.60	0.10	0.21
Fe^{2+}[mgL^{-1}]	0.50	1.50	1.61	0.22	0.01	n.a.	0.01
HCO_3^-[mgL^{-1}]	125	129	134	134	140	146	159
Cl^- [mgL^{-1}]	9.7	12.7	10.6	17.0	17.5	4.30	14.3
SO_4^{2-}[mgL^{-1}]	13.1	11.7	28.2	10.6	12.8	15.6	13.5

[a] year of measurement; [b] n.a. – not analyzed.

Table 6
Calculated quality parameters and annual committed effective dose resulting from the absorption of radionuclides from the study waters for an adult with the assumption of the consumption of 730 L of water per year.

Water	MN-01	MN-02	MN-03	MN-04	MN-05	MN-06	MN-07	MN-08	MN-09
D (mSv/year)	0.005	0.025	0.023	0.720	0.004	0.017	0.117	0.079	0.005
SAR	0.18	0.55	2.09	21.9	7.14	0.27	2.10	7.96	32.5
Na (%)	7.69	6.86	19.3	70.2	89.7	12.4	19.7	87.4	77.6

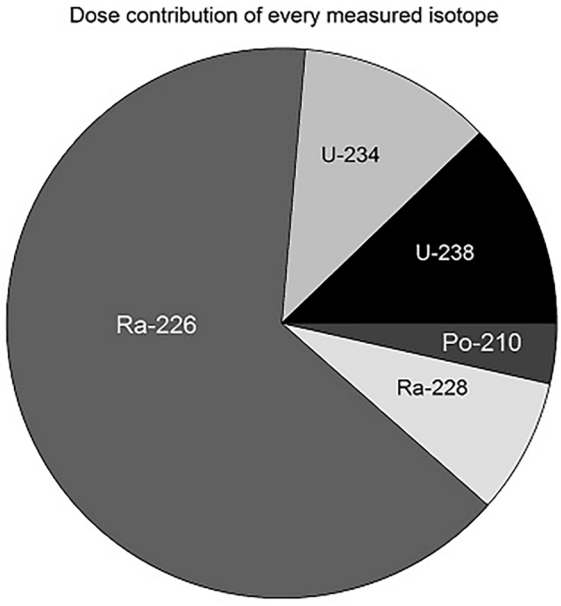

Fig. 11. Average contribution of each of the isotopes measured to the effective dose received.

have used thermal waters for therapeutic purposes. Therapeutic treatments are developing more and more and even the mud from the thermal water sources has been used in therapeutic bathing. In Vietnam, groundwater is considered thermal if the temperature of the water output is higher than 30 °C. Generally, the heat source for the thermal water is the geothermal gradient, though some underground water could be heated by magma fluid (water from My Lam MN-05). The average geogradient calculated using the temperature data of the studied thermal waters in North Vietnam is around 3.8 ± 0.5 °C per 100 m. All of the thermal waters studied originate from infiltration water, which was confirmed by both the analysis of the correlation between $\delta^{18}O$ and $\delta^{2}H$ isotopes and the calculated values of hydrochemical indices. The analysis of the major chemical ions pointed out that in the waters from Kim Boi – Hoa Binh (MN-01), Thanh Thuy – Phu Tho (MN-02), My Lam – Tuyen Quan (MN-05), Ban Moong – Son La (MN-06) and Tien Lang – Hai Phong (MN-08) there is a presence of infiltration water from the Miocene epoch. Both very high mineralization and very high radium concentration as well as low value of the rNa^+/rCl^- index in the Keng Ga thermal water (MN-04) indicate that the water exchange is limited in this source, the reservoir is isolated and fresh water inflow is restricted. The other studied groundwaters have a good hydraulic contact with infiltration waters. The lighter stable isotopes in the thermal waters: MN-02, MN-03, MN-05, MN-05 and MN-07 could be related to the both far distance from the water resources to the sea and their highland localization. While the relatively heavier stable isotopes in the rest studied waters: MN-04, MN-08 and MN-09 are possibly related to the closeness to the sea or/and lowland localization. High radium concentrations and analysis of the hydrochemical indices in the waters from MN-04, MN-07 and MN-09, suggest intensive interaction between the waters or/and a their long residence time in the water host formation as well as limitation in the exchange with modern waters. In the all of the studied waters the absence of ^{228}Ra or its activity concentrations significantly lower than those of ^{226}Ra suggest that the limestone is prevailing in the host formation. Excluding the waters from the Keng Ga (MN-04) and It Ong (MN-07), the linear positive relationships between ^{238}U and ^{226}Ra in remaining waters is observed. This phenomena can be attributed to the redox conditions (oxidising), which is prevailing in fractures and karst caves where the waters occur. The effective committed dose can be neglected if the studied waters are used in medical treatment or relaxation, but in the case of using the waters for drinking purposes, the dose could be much higher than the permissible value of 0.1 mSv/year even for the adult group. The contribution of the ^{226}Ra is above 70% in the total effective doses and it is a higher contribution compared to the contribution of the other isotopes analyzed in the waters studied. Furthermore, in several waters Keng Ga (MN-04), It Ong (MN-07) and Quang Hanh (MN-09) the uranium concentration (^{238}U) is higher than 15 μgL^{-1}. Such a high uranium content can accumulate in the kidneys and be of great danger to the human organism. In the case of irrigation purpose the waters MN-04 and MN-09 are classified as unsuitable, whereas the rest of the waters can be attributed to the excellent category.

CRediT authorship contribution statement

Nguyen Dinh Chau: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **Katarzyna Wątor:** Methodology, Formal analysis, Investigation, Writing – review & editing. **Piotr Rusiniak:** Methodology, Formal analysis, Investigation, Writing – review & editing. **Zbigniew Gorczyca:** Formal analysis, Investigation. **Duong Van Hao:** Formal analysis, Investigation.

Declaration of Competing Interest

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

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