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Naturally occurring radioactive materials (NORM) in the groundwater of two islands with various geologic settings in South Korea

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Abstract

Since 2007, Naturally Occurring Radioactive Materials (NORM) such as uranium-238 and radon-222 etc. in groundwater from the Community Water-supply Systems (CWS) in two islands have been studied in South Korea. In 71 samples from Ganghwa (G) Island, the maximum value of uranium-238 concentration is 72.21 μg/L. 3 CWSs (4.2%) exceeded the Maximum Contaminant Level (MCL) of 30 μg/L for uranium-238. The maximum value of radon-222 activity is 614 Bq/L. 28 CWSs (39.4 %) did not meet the United States Environment Protection Agency (US EPA) proposed Alternative Maximum Contaminant Level (AMCL) of 148 Bq/L for radon-222. At all CWS that did not meet the US EPA's MCL or AMCL, some appropriate actions were taken such as water treatment, alternative well development, mixing water of different origins, and so forth. In the 52 CWSs of Jeju (J) Island, the maximum value of uranium-238 and radon-222 concentrations are 1.37 μg/L and 94.83 Bq/L, respectively. All values for uranium-238, gross alpha, and radon-222 meet MCL and proposed AMCL of US EPA drinking water standard. The two islands have different geological settings that are believed to be the causes of the big differences in the NORM levels. Geologically an old island has much higher NORM values than a young island formed in the Quaternary Period due to hydrogeological factors such as recharge and infiltration rates of precipitation. The residence times in the aquifers for water-rock (mineral) interactions are very different from each other.

Keywords: geologic setting, groundwater, uranium-238, radon-222

1. INTRODUCTION

In South Korea, some community water-supply systems with high levels of uranium-238 and radon-222 in drinking roundwater have been reported and broadcast several times by major media such as News 9 by the Korean Broadcasting System (KBS) and Newsdesk by Munhwa Broadcasting Corporation (MBC)

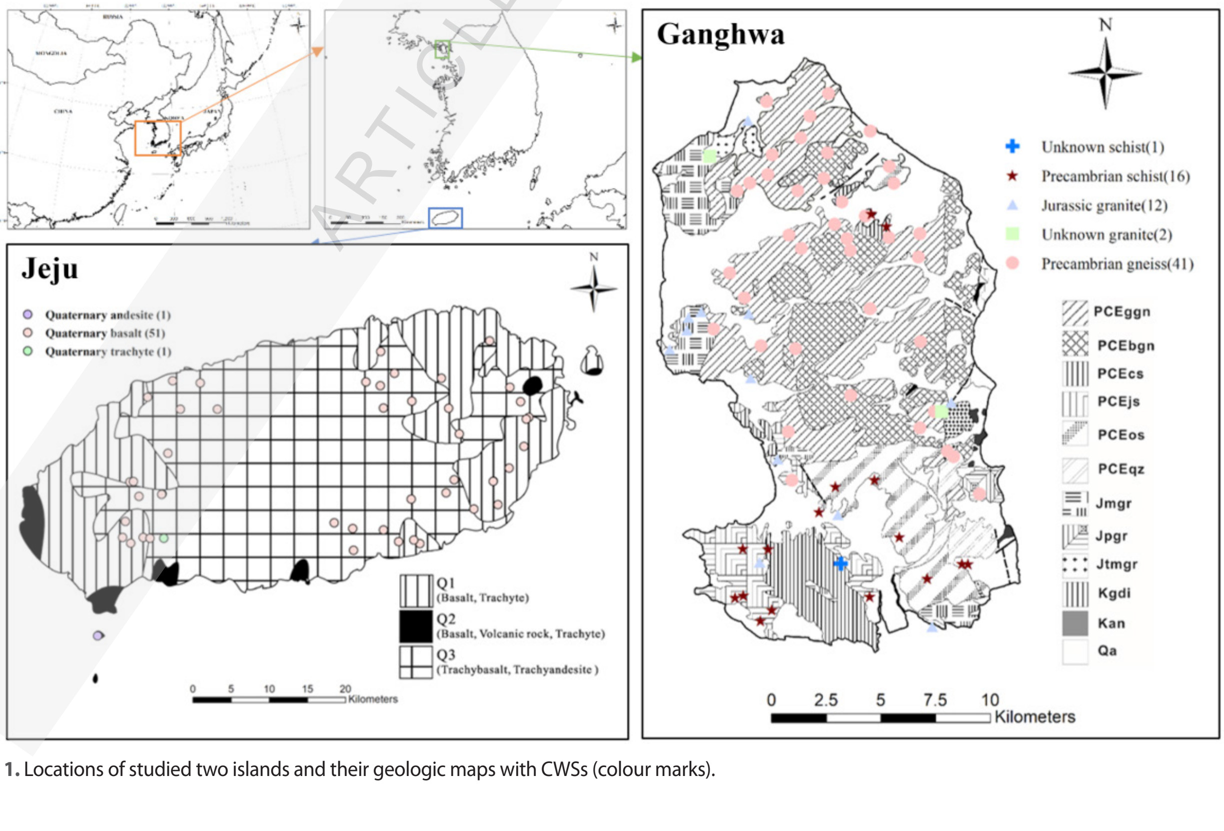

Figure 1. Locations of studied two islands and their geologic maps with CWSs (colour marks).

Television News etc, and the various newspapers such as The Hankyoreh and The Chosunilbo etc., since 1998. The news made the national assembly of the republic of Korea, requiring several actions by Ministry of Environment during the inspection and investigation of state administration. Studies and investigations, therefore, on naturally occurring radioactive materials (NORM) in groundwater have been performed since 1999. Uranium-238 is known as the chemical that is increasing kidney toxicity (US EPA, 2000) and radon-222 is infamous as the NORM chemical causing lung cancer (National Academy Press, 1999).

This study aims to determine the levels of the NORM in groundwater from the community water-supply systems (CWSs) on two islands in South Korea (Fig. 1). Areas, population, and annual precipitations of two islands are 410.6 km^2, 68,010 people, and 1,076 mm/year in 2016 for Ganghwa (G) Island and 1848.7 km^2, 641,597 people, and 1,416 mm/year in 2016 for Jeju (J) Island (KOREA LAND & HOUSING CORPORATION, 2017).

The studied islands have different geological settings investigated and mapped by the Korea Institute of Geoscience and Mineral Resources (KIGAM). The G island is composed of Precambrian metamorphic rocks, Jurassic Daebo granite, Cretaceous Bulguksa granitoids, Cretaceous acidic dykes and Quaternary deposits. The metamorphic rocks of G island consist of quartz, K-feldspar, plagioclase, biotite, moscovite and hornblende with rare garnet. The granitoids consist of quartz, plagioclase, K-feldspar, biotite, hornblende and zircon, two-mica granite. The acidic dykes are classified into felsite dyke, rhyolite dyke, pegmatite vein and quartz vein (HWANG & KIHM, 2005). The J island is a shield volcano, composed mainly of basaltic lava flows and subordinate amounts of pyroclastic and sedimentary rocks. The lavas of the Jeju island are known as basalt, trachybasalt, basaltic trachyandesit, trachyandesite and trachyte formed in pre-Pliocene (Cretaceous) volcanic and plutonic rocks. Volcanic rocks of J island are composed of olivine, plagioclase, and pyroxene (PARK et al., 2000).

The NORM concentrations in the aquifers of the two islands with various geological settings are evaluated.

2. MATERIALS AND METHODS

In order to define the levels of NORM for uranium-238 and radon-222 in groundwater from G island located in the north-western side and from J island located in the southern part of South Korea, a total of 123 CWSs using groundwater as a drinking water resource had been investigated for 10 years from 2007 to 2016 (NIER-2007, -2008, -2009, -2010, -2011, -2012, -2013, -2014, -2015, -2016).

Groundwater samples for uranium-238 were filtered by 0.45 μm membrane (Advantec®) in the field and transported to the laboratory and then analysed by Inductively coupled plasma - mass spectrometer (ICP-MS, Agilent 7500 Series and PerkinElmer NexIONTM 350D) in the National Institute of Environmental Research (NIER). The range of uranium-238 detection limits (DL) measured every year for the quality assurance and quality control (QA/QC) was from 0.01 to 0.09 μg/L. The range of accuracies and precisions for the QA/QC were 88.85% to 99.67% and 0.63% to 4.92%, respectively. Samples for radon-222 were prepared in the field. 8 mL of the groundwater was injected into a 20 mL high density polyethylene vial (HDPE) to which 12 mL of scintillation cocktail (Optiphase Hisafe3) had been added. The vial was shaken for about 5 minutes to mix the sample with the cocktail. The sample was then equilibrated for 3 hours and then counted for 60 minutes in the liquid scintillation counter (LSC, PerkinElmer Quantulus 1220) using energy discrimination for alpha particles in NIER (HAHN & PIA, 1991). One litre of groundwater was sampled and tested for gross alpha, with 15 mL of 1N HNO_3 added to the samples on-site prior to transfer to the laboratory. The appropriate amount of sample was collected for analysis. Collected samples were evaporated, concentrated and dried to produce solid samples. The solid samples were analyzed by gas-flow proportional counter (GPC, CANBERRA Tennelec™ Series 5) in NIER (US EPA, 1980). The range of minimum detectable activities (MDA) calculated every year for the QA/QC is from 0.02 to 0.04 Bq/L.

3. RESULTS

In 71 groundwater samples from G island, the maximum and minimum values of uranium-238 concentrations were 72.21 μg/L and ND (not detected; below the detection limit), respectively. The average and median values of uranium-238 in the samples were 5.56 μg/L and 1.32 μg/L, respectively. 3 CWSs (4.2%) exceeded the maximum concentration level (MCL) of 30 μg/L for uranium-238, which is regulated by the national primary drinking water standard of the United States Environmental Protection Agency (US EPA). The maximum and minimum values of radon-222 activity were 614 Bq/L and 6 Bq/L, respectively. The average and median values of radon-222 in the groundwater were 133 Bq/L and 111 Bq/L, respectively. In 28 CWSs radon-222 activity concentrations exceeded the AMCL value of 148 Bq/L. The maximum and minimum values of gross alpha activities were 0.51 Bq/L and ND (not detected; below the minimum detectable activity), respectively. The average and median values of gross alpha in groundwater were 0.07 Bq/L and 0.04 Bq/L, respectively. All gross alpha values meet the maximum concentration level (MCL) of 0.55 Bq/L, regulated by the US EPA's national primary drinking water standard. All CWS that did not meet the US EPA's MCL or AMCL have taken appropriate action, such as water treatment, alternative well development, mixing water of different origins, and so forth.

In the 52 CWSs of J island, the maximum and minimum values of uranium-238 concentrations were 1.37 μg/L and ND (not detected), respectively. The average and median values of uranium-238 in the groundwater were 0.15 μg/L and 0.07 μg/L, respectively. The maximum and minimum values of radon-222 activities were 94.83 Bq/L and 0.07 Bq/L, respectively. The average and median values of radon-222 in the groundwater were 23.74 Bq/L and 18.32 Bq/L, respectively. The maximum and minimum values of gross alpha activities were 0.24 Bq/L and ND, respectively. The average and median values of gross alpha in the groundwater were 0.03 Bq/L and 0.00 Bq/L, respectively. All values for uranium-238 and gross alpha meet the maximum concentration levels (MCL) regulated by the US EPA drinking water standard of 30 μg/L and 0.55 Bq/L, respectively. None of the radon-222 concentration values exceeded the US EPA proposed alternative maximum concentration level (AMCL) of 148 Bq/L (Fig. 2). Figure 3 shows the box plots of uranium-238, radon-222, and gross alpha values of groundwater samples from the two islands. Table 1 (NIER-2007, -2008, -2009, -2010, -2011, -2012, -2013, -2014, -2015, -2016) shows the uranium-238, radon-222, and gross alpha concentrations of minimum, maximum, average and median values. In addition, this table includes the basic statistical values of well depths. However, we could not acquire the data of all screen depths and some of the well depths of two islands. The number of well depth values is 48 from the G island. The number of well depths is 45 from the J island. The ranges of

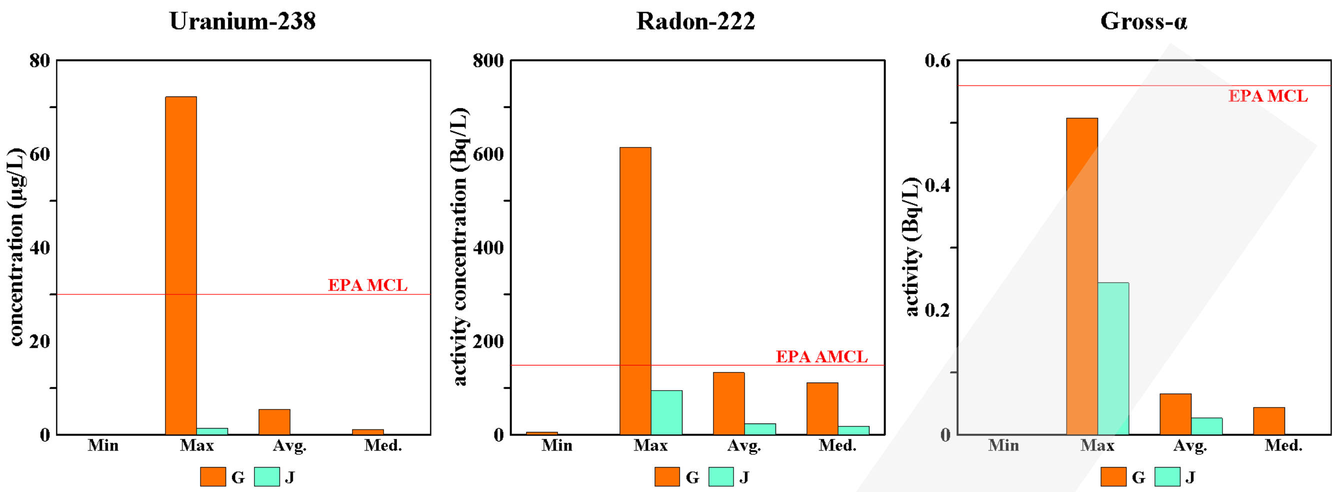

Figure 2. Bar charts of uranium-238, radon-222, and gross alpha values of CWSs in two islands.

Table 1. Statistics of well depth, uranium-238, radon-222, and gross alpha values.

Site	Basic Statistics	Well depth (m)	Rn-222 (Bq/L)	Uranium-238 (μg/L)	Gross-α (Bq/L)
G island (n=71)	Min.	70	6.2±0.3	ND*	ND**
	Max	250	615±3	72.21	0.51
	Avg.	98	132.78	5.49	0.07
	Median	100	111.01	1.16	0.04
J island (n=52)	Min.	43	0.07±0.02	ND*	ND**
	Max	450	94.8±1.5	1.37	0.24±0.16
	Avg.	182	23.74	0.15	0.03
	Median	143	18.32	0.07	0.00***

* ND of uranium-238 : below the detection limits (DL, 0.01 ~ 0.09 μg/L)
** ND of gross-α: below minimum detectable activities (MDA, 0.02 ~ 0.04 Bq/L)
*** All values that were below the MDA assign the 0

well depths in the J island is much wider than those in the G island. It depends on the different geological settings of the two islands.

4. DISCUSSION

The two islands have different geological settings that may explain the causes of the big differences observed in the NORM levels. Most of the G island is composed of Precambrian metamorphic rocks, Jurassic granite, Cretaceous granitoids, Cretaceous acidic dykes and Quaternary deposits (HWANG & KIHM, 2005). J island is a shield volcano, consisting mainly of basaltic lava flows and subordinate amount of pyroclastic and sedimentary rocks formed in Pleistocene Epoch, Quaternary Period, Cenozoic Era (PARK et al., 1998).

Uranium, one of the immiscible elements, increases its content as the fractional crystallization process in which crystalline rock is produced. Thus, mafic rocks containing basalts contain little uranium (GREEMAN, 1991). The common ranges of uranium contents are 2.20 ~ 15.00ppm in granite, 0.10 ~ 10.00ppm in gneiss and 0.10 ~ 2.30ppm in basalt (DAHLKAMP, 2013). The rocks mainly composed of G island are granitoids and metamorphic rocks, and the rocks composed of J island are volcanic rocks. Porosity of crystalline rocks including granite and metamorphic rocks is < 0.001 to 0.01. Porosity of basalt is < 0.01 to 0.10 (INTERA ENVIRONMENTAL CONSULTANTS, INC., 1983). Due to different types of rocks, there is a large difference in transmissivity. The average transmissivity values of groundwaters measured in G island and J island are 5 m^2/d and 560 m^2/d, respectively (HAN RIVER FLOOD CONTROL OFFICE et al., 2012). The recharge and infiltration rate of precipitation in J island is much higher than that of G island, and the span of water cycle from recharge into aquifers to discharge from the aquifers in J island is much shorter than that in G island. The average electrical conductivity of the national groundwater network in G island is 449.49 μS/cm and that in J island is 126.51 μS/cm (NATIONAL GRONDWATER INFORMATION CENTER). The electrical conductivity

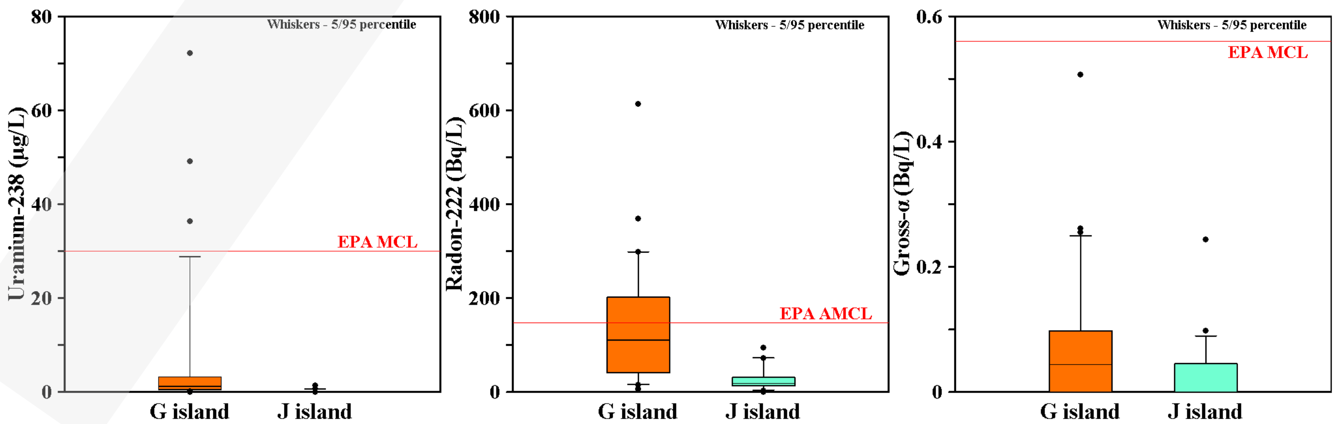

Figure 3. Box plots of uranium-238, radon-222, and gross alpha values of CWSs in two islands.

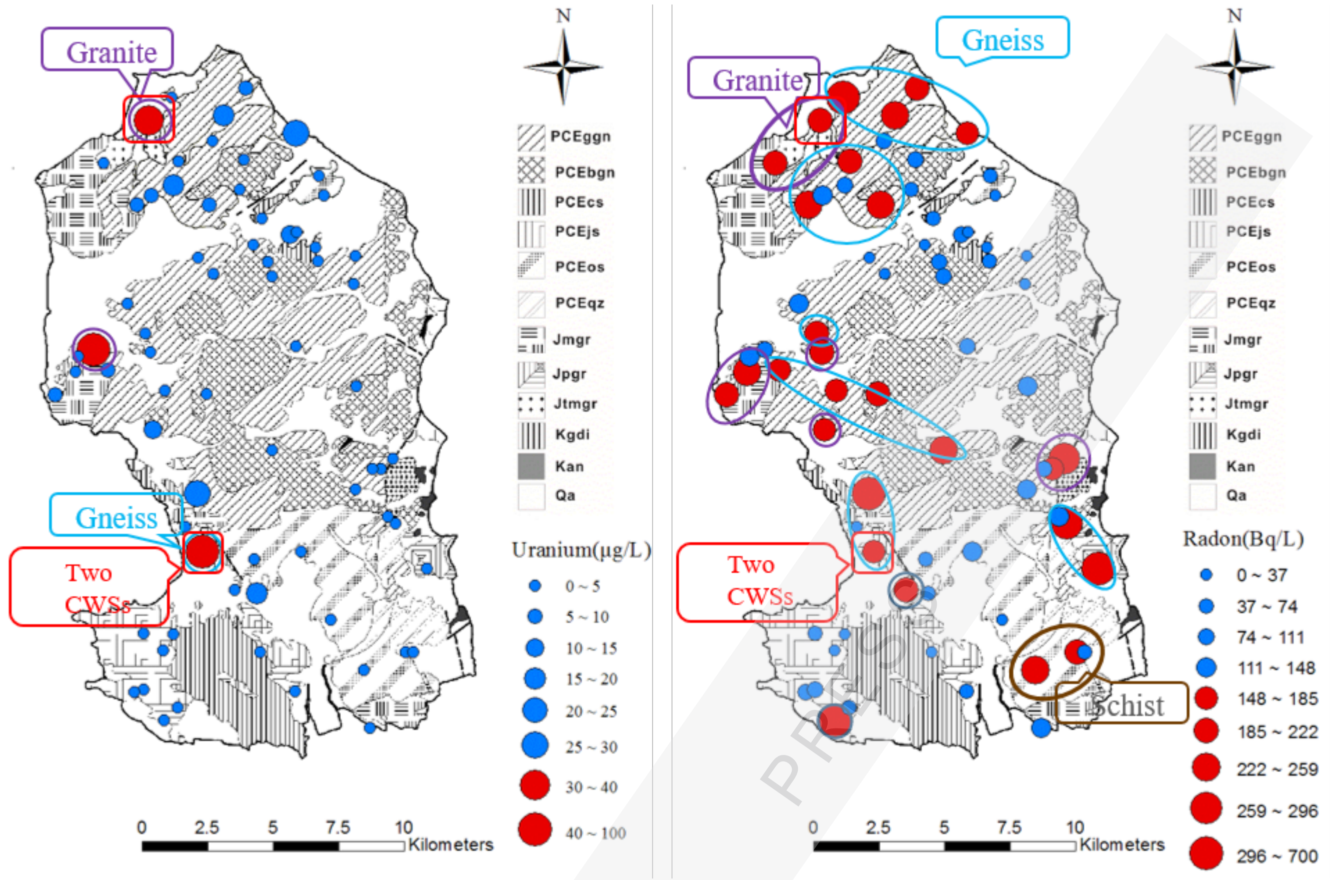

Figure 4. Bubble plot of uranium and radon concentrations in the G island.

can be converted to total dissolved solids (TDS). The higher the TDS value, the longer the residence time is expected because high TDS values are the result of long-term water-rock reactions (NELSON, 2002). The difference of electric conductivities indicates that the groundwater in the aquifers of G island with a long residence time has much higher the NORM levels than that of J island with its relatively short residence time. This indicates that groundwater in metamorphic and granitic rocks with relatively longer water-rock interaction times has a much higher level of uranium, radon, and gross alpha values than basaltic rocks with relatively short water-mineral interaction times.

Correlation coefficient and p value between uranium-238 and radon-222 concentrations were 0.22 and 0.058 in the G island, respectively. There is little correlation between uranium-238 and radon-222 concentrations in groundwater in the G island. This relationship may be attributed to their unique physico-chemical characteristics such as half-lives (^{238}U: 4.5 billion years, ^{222}Rn: 3.82 days), phases (^{238}U: dissolved ions, ^{222}Rn: inert gas), etc. Groundwaters discharged from Precambrian biotite gneiss, Jurassic biotite granite areas and Jurassic two-mica granite areas contain relatively high values of uranium-238 concentrations (>30 μg/L). Groundwaters bearing relatively high levels of radon-222 activity (>148 Bq/L) are dominantly found in the regions of Precambrian granitic gneiss. Groundwaters discharged from nine geologic settings that are Jurassic biotite granite, Precambrian biotite gneiss, Precambrian quartz schist, Precambrian mica schist, Precambrian biotite schist, unknown granite, Jurassic granite, Jurassic two-mica granite, Jurassic porphyritic biotite granite areas have also relatively high levels of radon-222 activities (>148 Bq/L) (Fig. 4). No values of gross alpha exceed the MCL of US EPA.

2 and 8 CWSs (64.3 %) exceeded US EPA MCL/AMCL values of uranium-238/radon-222 among 14 CWSs in granitic

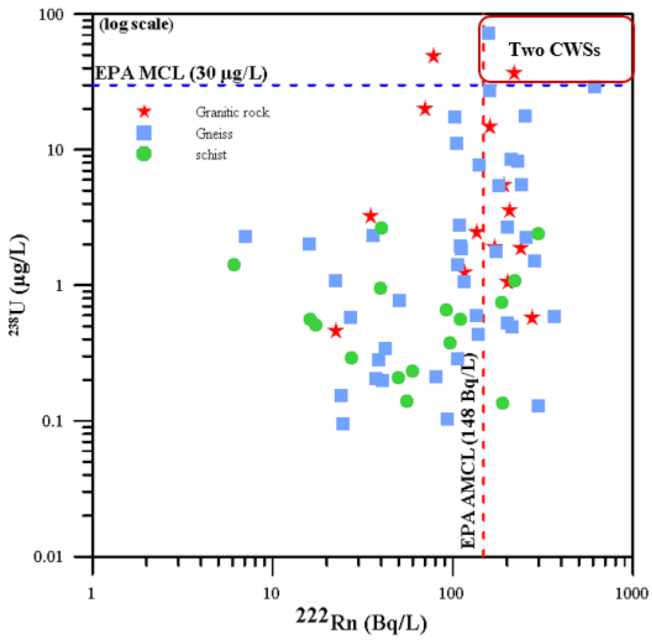

Figure 5. Uranium-238 and radon-222 values based on geological settings in the G island.

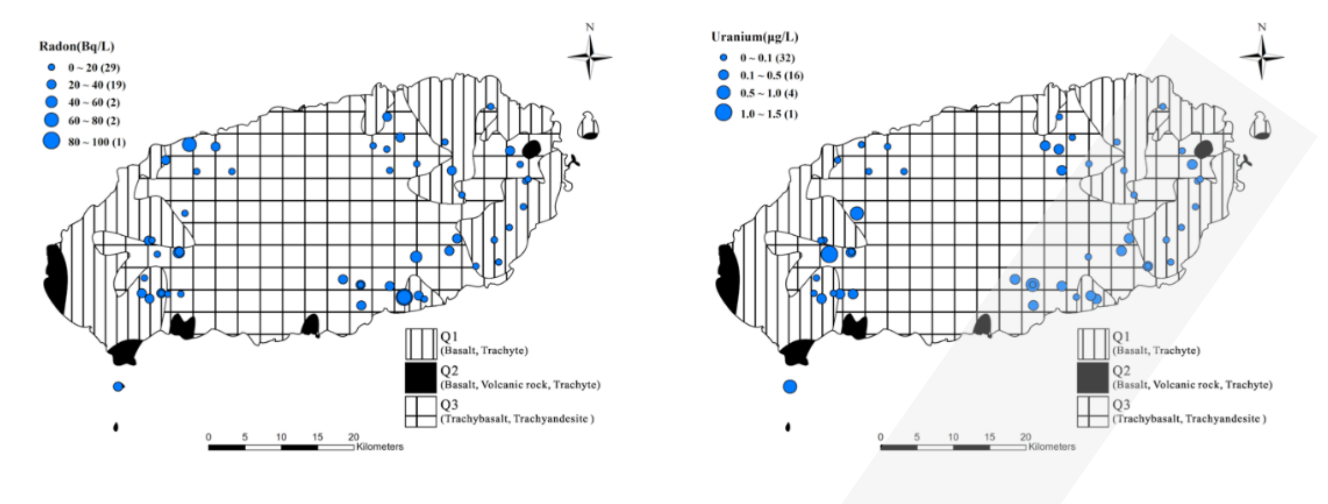

Figure 6. Bubble plot of uranium-238 and radon-222 concentrations in the J island.

rock areas, respectively. 1 and 16 CWSs (39.0 %) exceeded US EPA MCL/AMCL values of uranium-238/radon-222 among 41 CWSs in gneiss rock areas, respectively. 4 CWSs (23.5 %) exceeded US EPA AMCL proposed value of radon-222 among 17 CWSs in schist rock areas (Fig. 5).

In the J island, uranium-238 and gross alpha values in groundwater are very low, however, radon-222 values show their existence in the groundwater system. This implies that a shield volcano island formed in the Quaternary Period has little NORM in their aquifers (Fig. 6). The highest concentrations of radon-222 (94 Bq/L) and uranium-238 (1.37 μg/L) in groundwater are found in Q3 consisting of Trachy-basalt and Trachy-andesite.

5. CONCLUSION

NORM in groundwater from the two islands with different geological settings in South Korea had been studied from 2007 to 2016. Jeju Island has low NORM values under the MCL and AMCL values of uranium-238, gross alpha, and radon-222. However, Ganghwa Island has relatively high values of NORM with 3.2 % and 39.4 % of uranium-238 MCL and radon-222 AMCL exceeding ratios, respectively. The result indicates that the geology of aquifers is the major factor of the NORM contents of groundwater.

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