Natural Uranium Distribution at the KURT Site

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Contents

Current Issue- Key Recommendations from PECOS

- **Public Engagement Commission on SNF Management (PECOS)**
  - **The non-government commission, to gather various opinion on SNF management**

Key Recommendations (in 2015)

- **The government should *operate the permanent disposal facilities by 2051*.**
- **A candidate site, in which both of *URL facility* and centralized *interim storage facility* to be constructed, has to be selected by 2020. Demonstration and verification programs for disposal concept, technology and its safety at the URL should start by 2030.**
- **SNF could be stored in on-site dry-type storage facility prior to transferring to centralized interim storage facility.**
- **Priority on SNF issues including storage, transportation, disposal and pyro-processing to minimize volume and toxicity of waste should be evaluated and systematic plans for each issue should be established**
- **Establishment of a new public corporation (e.g. SNF technology and management corporation) for safety, responsibility, efficiency and transparency of SNF management**
- **Enactment of special law on SNF management**

Current Issue- Basic plan from MOTIE in 2016

❑ MOTIE announced a Basic plan for HLW management (July, 2016)

- ❖ To select one site for a site specific URL, Centralized interim storage facilities, and a HLW repository
  - ***Site selection (+ 12 yrs)***
  - ***Centralized interim storage facilities construction (+7yrs)*** + ***Site specific URL construction and operation (+14yrs)***
  - ***HLW repository construction (+10yrs)***
- ❖ To consider international cooperation on sharing storage & repository for HLW management
- ❖ To achieve key technologies punctually for safe and economic HLW management
- ❖ To keep transparency in HLW management and communication with publics

❑ Currently, a special law for HLW management is to get approved by the Korean National Assembly

Introduction

Introduction- Background I

❑ Uranium (U) Analogue

- U is a **major element among naturally occurring radioactive materials in rocks** due to its abundance in subsurface environments. High U contents are mainly found in granite, phosphate, and organic-rich black shales.
- The **distribution of U and understanding of its migration and retardation processes are very important in terms of geological disposal of radioactive wastes** as radionuclides released from radioactive waste repository might be migrated and retarded in subsurface environments.
- The **ubiquitous presence of U in rocks and groundwaters** makes it an **ideal natural analogue for studying geochemical behavior of radionuclides** in a deep geological repository for final disposal of HLW.

One of important issues in the deep geological disposal of high-level radioactive waste is the **confident prediction of natural processes** during time scales up to hundreds of thousand years. (Suutarinen et al., Rdiochim. Acta 52/53, 1991)

➔ One possibility of resolving this issue is to study the **migration phenomena using naturally occurring radionuclides** as analogues of radioactive waste

Introduction- Background II & objectives

❑ Uranium (U) Isotopes

- Three isotopes: ^{238}U (4.5×10^{9} y), ^{235}U (7.1×10^{8} y), ^{234}U (2.5×10^{5} y)
- Mass percentages: ^{238}U (99.27%), ^{235}U (0.72%), ^{234}U (0.006%)
- Due to long radioactive half-lives, **^{238}U and ^{234}U isotopes are in secular equilibrium** in all minerals and rocks older than one million years in a closed system or undisturbed minerals since ^{234}U is a daughter product of ^{238}U. (Tripathi et al., J. Radioanal. Nucl. Chem. 295, 2013)
- Variations in AR have been used as a sensitive chemical indicator for identifying isotopically distinct groundwaters and studying geochemical processes

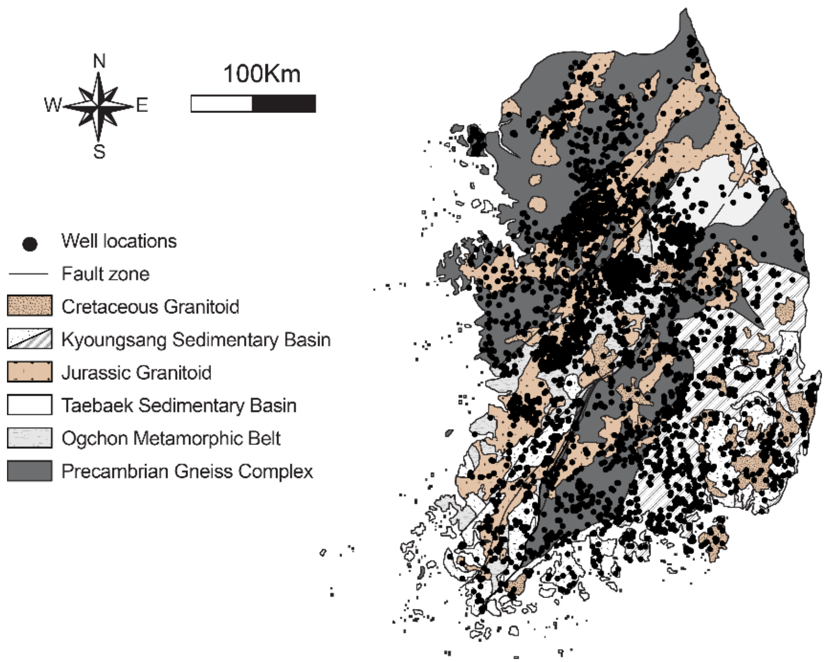
  → Information for **origin of U, groundwater redox conditions, groundwater-rock interaction, and mobility and retardation of U**

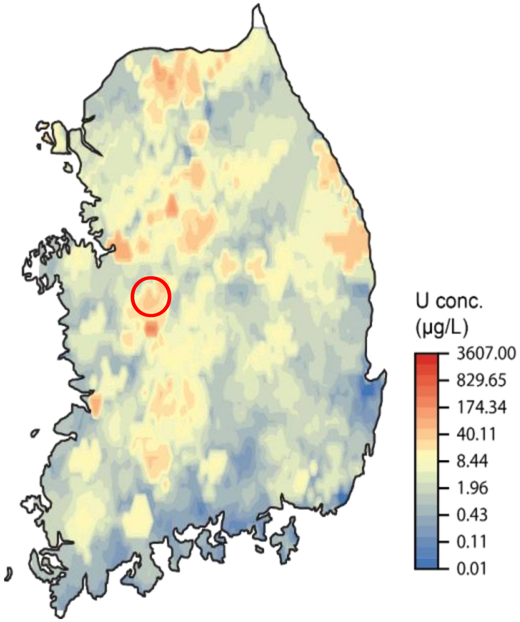
Introduction- objectives

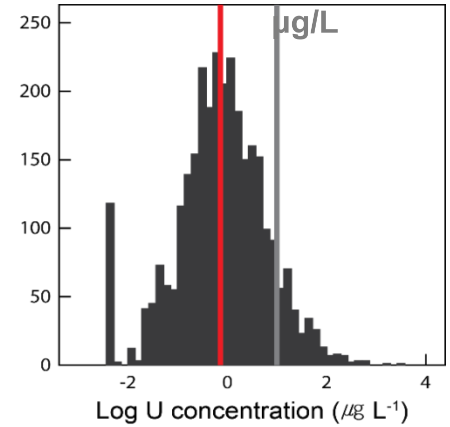
❑ Specific objectives of this study are

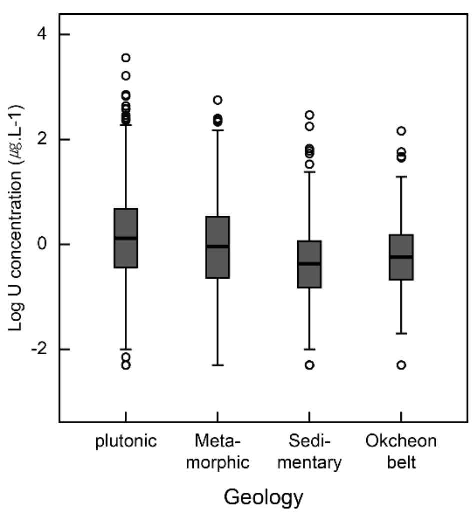
- To determine the environmental ***distribution of natural uranium*** in groundwater and its ***geological relationship***
- To understand the ***potential migration pathway*** of natural uranium in fractured bedrock aquifer through excavation and hydraulic test to water conductive feature
- To estimate ***uranium species*** and concentration in the granitic groundwater using TRLFS (Time-Resolved Laser Fluorescence Spectroscopy) and model calculation
- To investigate ***geochemical behaviors of uranium*** using the activity ratios of $^{234}U/^{238}U$.
- **The main goal of this study is to evaluate the distribution of uranium and the understanding of its potential migration and retardation processes in granitic subsurface environments.**

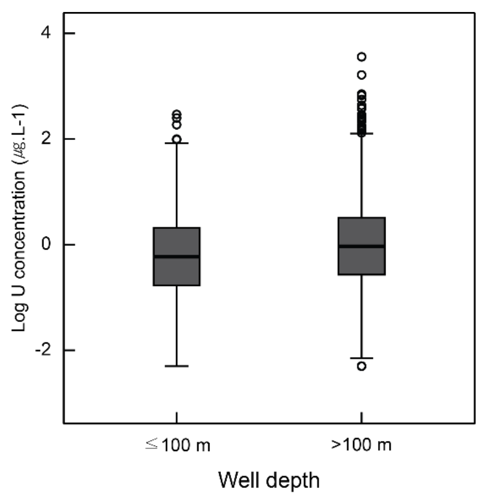
Groundwater Uranium in Korea











	N	mean	min.	med.	max.	unit
This study	3009	8.11	<0.01	0.79	3,607	μg/L

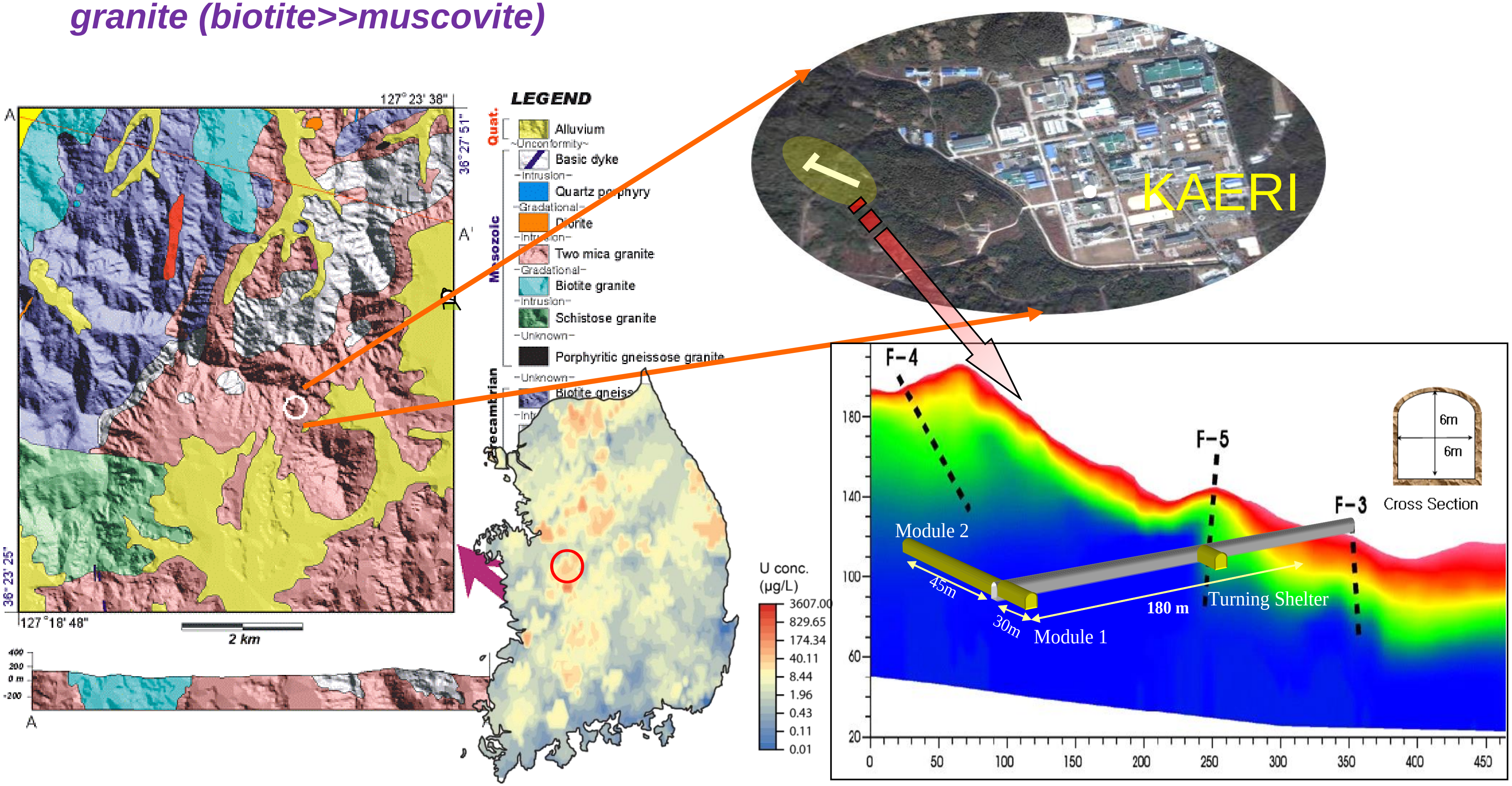
Geology	N	Mean	Min.	Med.	Max.	Stdev.
plutonic	1312	13.15	0.01	1.31	3607.00	115.90
metamorphic	646	7.55	0.01	0.90	563.56	31.58
sedimentary	849	1.95	0.01	0.43	293.00	12.48
okcheon belt	202	2.99	0.01	0.57	145.20	11.99

Well depth	N	Mean	Min.	Med.	Max.	Stdev.
=<100 m	936	4.20	0.01	0.59	293.00	17.22
>100 m	2073	9.87	0.01	0.93	3607.00	93.69

(not published)

Study site- KAERI site

- Study site : **KAERI & KURT** (*KAERI Underground Research Tunnel*)
- Geology around KAERI site : *Pre-Cambrian metamorphic rocks* (biotite gneiss and banded gneiss, limesilicate, biotite shist), *Mesozoic plutonic rocks* (schisotse graite, biotite granite, two mica granite), dykes
- KURT located in western part of KAERI site : ***medium to coarse-grained Jurassic two-mica granite (biotite>>muscovite)***



Study site- Overview of KURT & its Extension

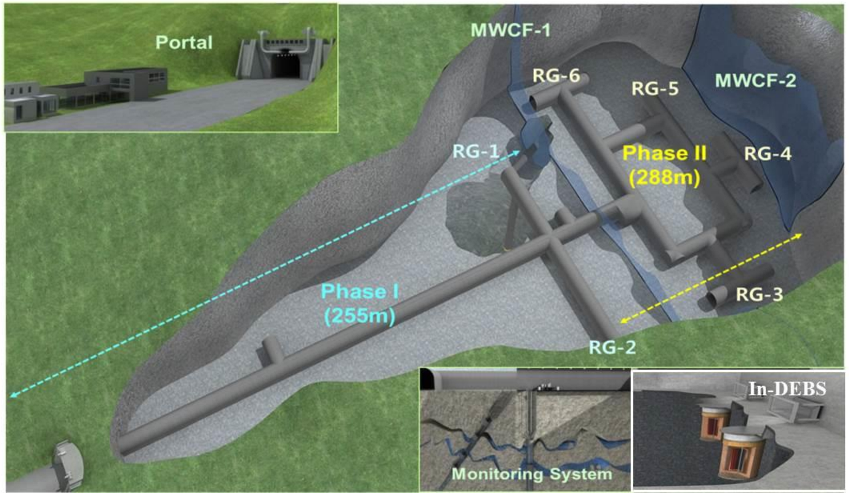
KAERI Underground Research Tunnel (KURT)

- **To improve geoscientific investigation technologies for the site characterization**
- **To provide the geoscientific data on the site scale for system development and safety assessment studies**
- **To develop the detailed site descriptive model on the site and block scales**

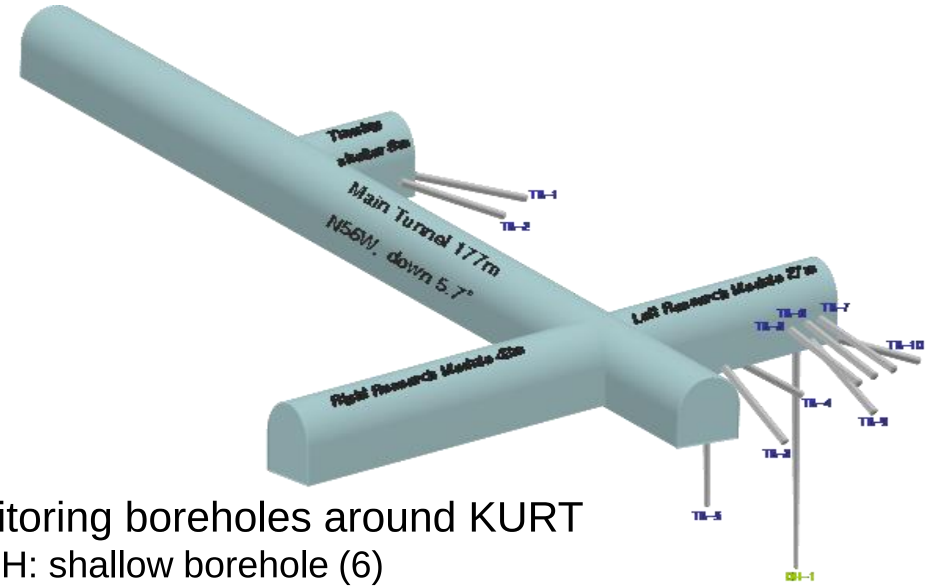
- Off-site generic URL, not site-specific URL
- R&D site, not potential site for waste disposal
- Purpose-built, not pre-existing excavation
- Crystalline rock basement (two-mica granite)
- No radioactive materials used

KURT = KAERI Underground Research Tunnel

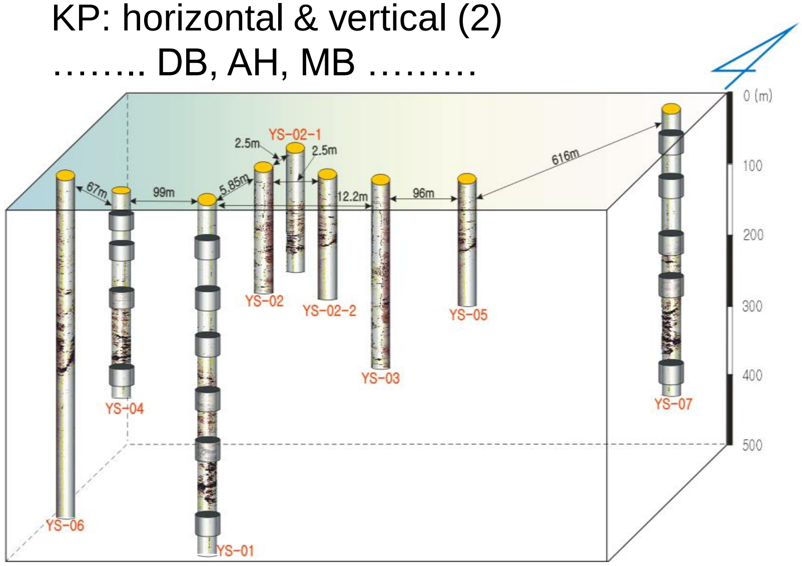
- Constructed in 2004 (Phase I) & Extended in 2015 (Phase II)
- Total length: 543 m
- Depth (from the peak): 120 m
- 6 research modules
- Slope: -10% (Phase I)

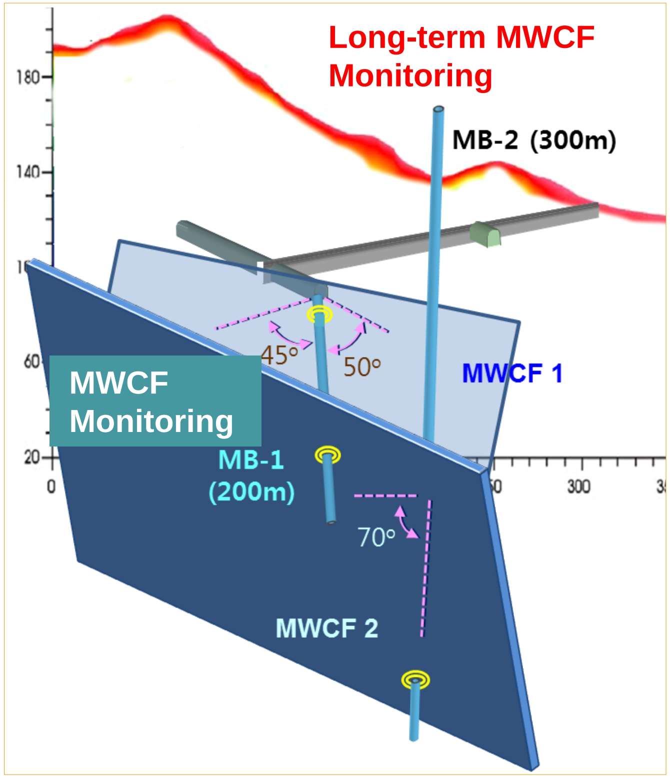


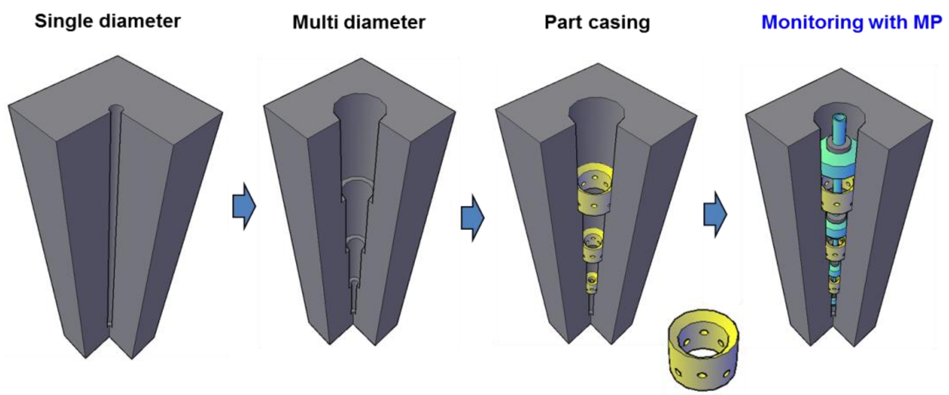
Monitoring Boreholes in KAERI



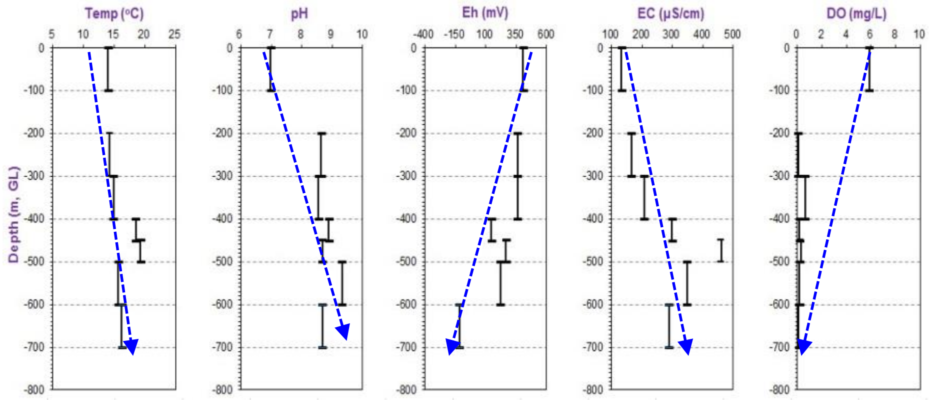
Monitoring boreholes around KURT
- BH: shallow borehole (6)
- YS: deep borehole (7)
- KP: horizontal & vertical (2)
- …….. DB, AH, MB ………

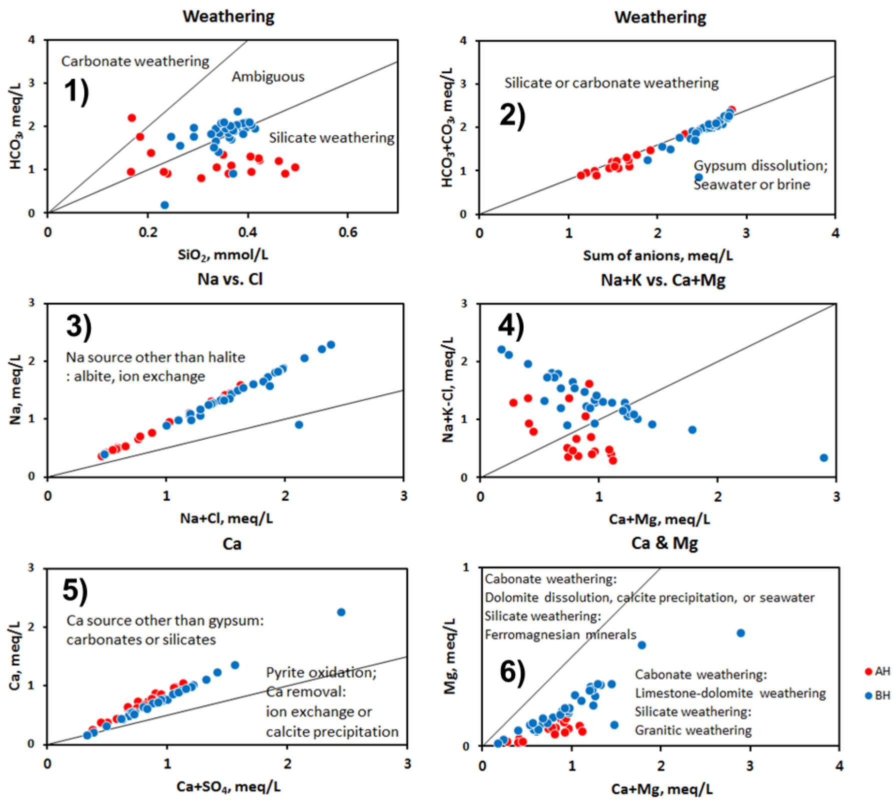






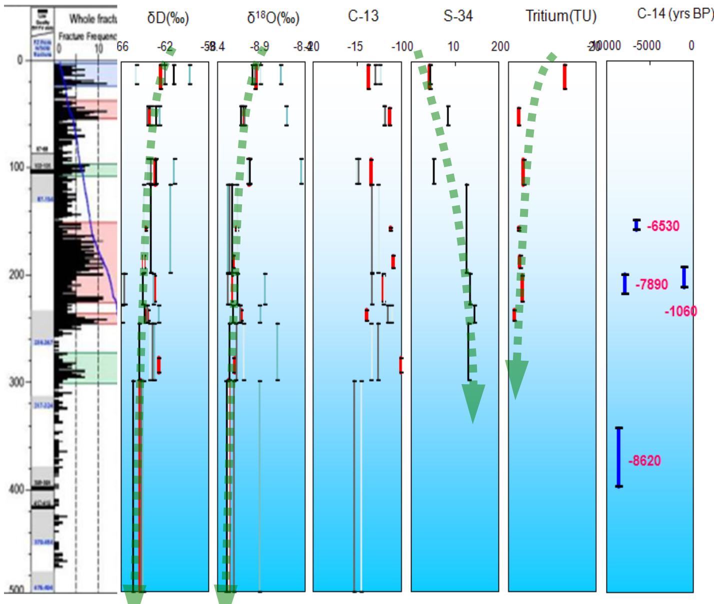
General features of KAERI GW (I)





With increasing depth

- **Increases : Temp., pH, EC, $\delta^{34}S$**
- **Decreases : Eh, DO, δD, $\delta^{18}O$, Tritium**



Water-rock interaction

: silicate weathering dominant(1)

: major anion (HCO_3+CO_3)(2)

: weathering of Na-bearing mineral(3)

: Ca+Mg vs. Na+K (reverse correlation)(4)

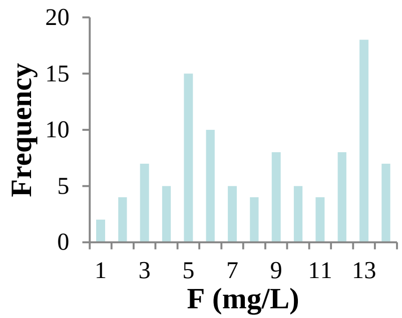
: low SO_4(5)

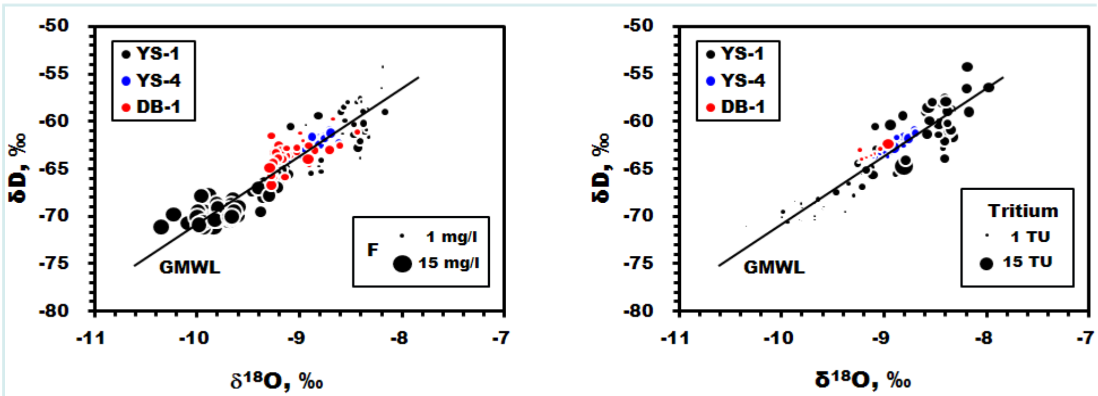
: trend of granite weathering(6)

General features of KAERI GW (II)

F distribution

Min	0.6
Max	13.7
Average	7.8
Median	7.8

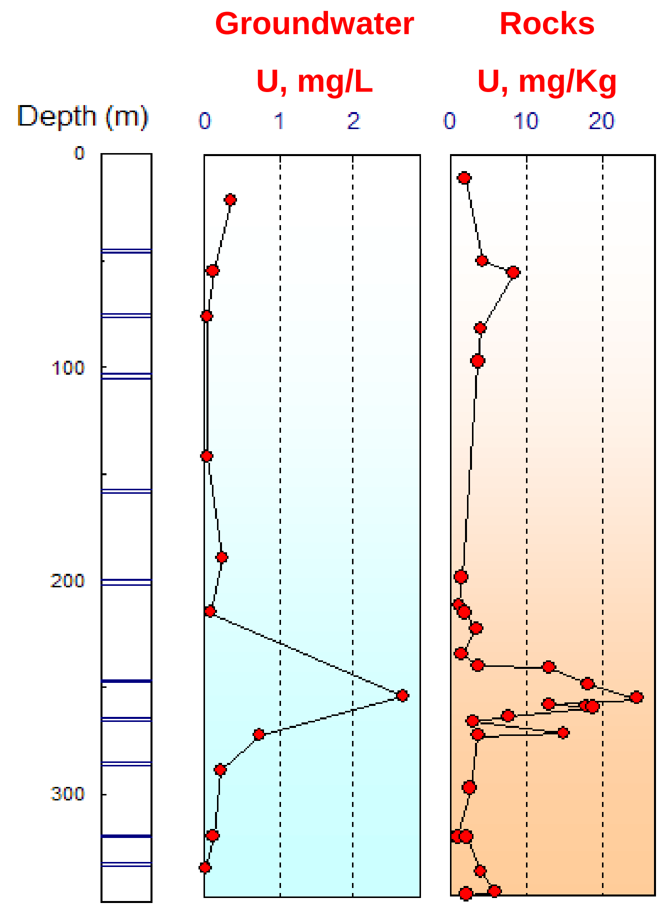




- ✓ **Good correlation between δ18O and tritium:** ***groundwater recharged at high elevation show the long residence time***
- ✓ **Negative relationship between ^3H and F:** ***groundwater with long residence time show high F concentration***
- ✓ ***F concentration in groundwater might be controlled by residence time***

U distribution

In borehole YS-4



- **In fracture zone (-250m)**
  - high U (25 ppm) of rocks
  - high U (3 ppm) of water
  - oxidized condition

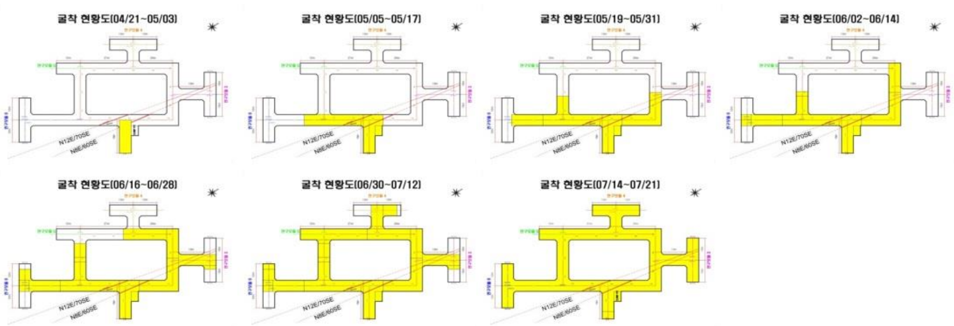
Hydrological and geochemical monitoring during KURT extension (in 2014)

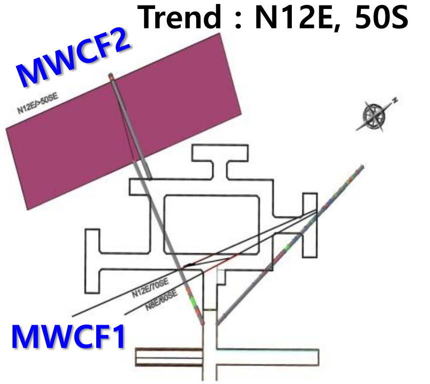
- Hydrological and geochemical connectivity

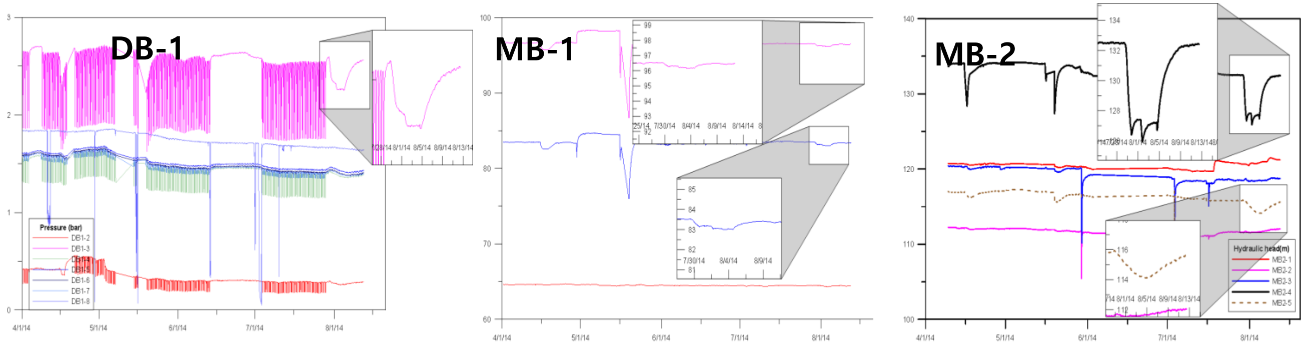
KURT Extension

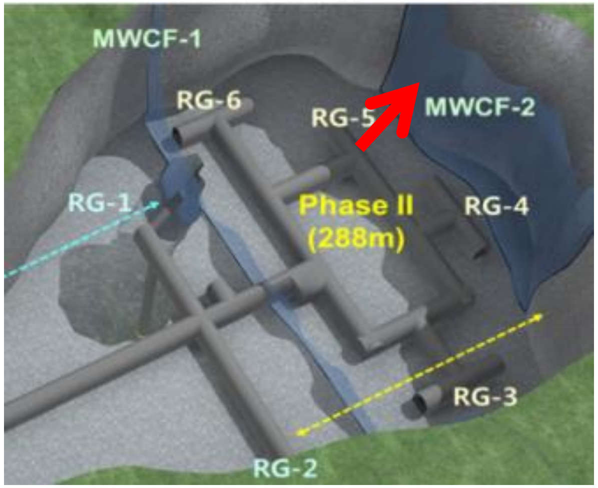
❑ KURT Extension (in 2014)

- MWCF (Major Water Conductive Feature)
- During extension period (4 months),
  - *hydraulic pressure changes were observed*
  - no significant variation of groundwater chemistry
- **MWCF2** targeting borehole and pumping (July 30th, 2014)
  - *hydraulic pressures in monitoring boreholes were decreased*
    : **DB-1 (i3), MB-1 (i2), MB-2 (i4)**
  - coincide the regions ***showing high U peaks***
    → MWCF2 is *main flow path of U*







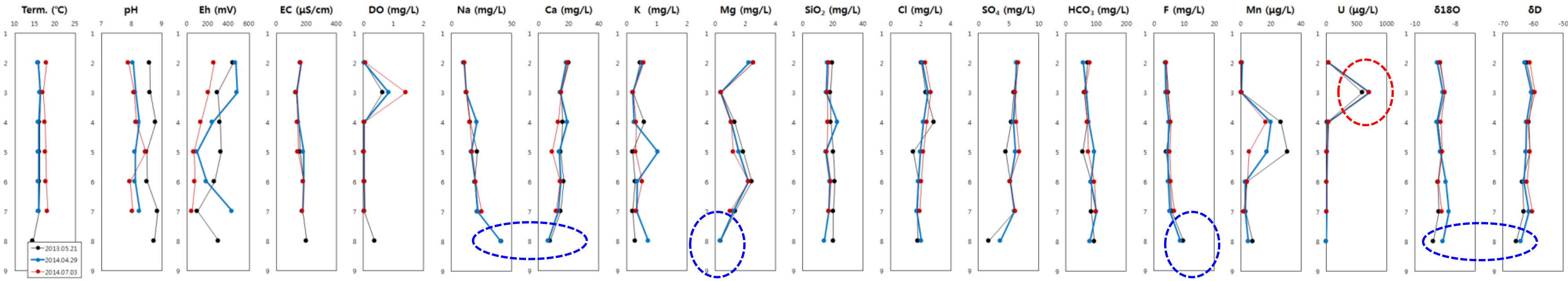


Geochemical monitoring

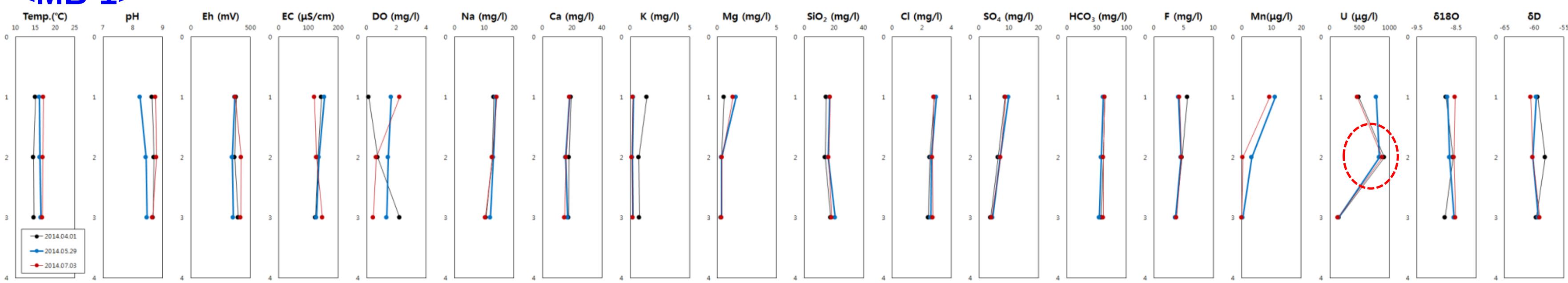
- High peak of U conc.
: *DB-1 (I3), MB-2 (I4), MB-1 (I2)*

- From GW Sampling Campaign (pre-/mid-/end-term excavation)

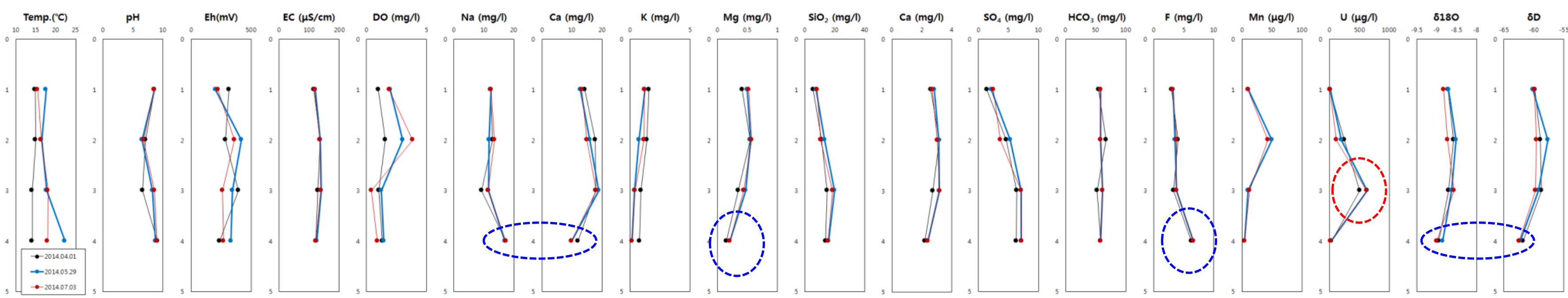
<DB-1>



<MB-1>



<MB-2>



Hydrochemical connectivity

- Hydrochemical Connectivity and physico-chemical parameters through 3 times monitoring in the boreholes monitored (DB-1, MB-1, and MB-2)

	DB-1							MB-1			MB-2			
	2	3	4	5	6	7	8	1	2	3	2	3	4	5
	43.5-59.5	92-116	156-159	183-194	201-226	234-244	279-293	0-68	68-120	120-200	75-112	120-140	150-200	250-300
Temp.	16.5±1.2	16.5±0.4	16.5±0.9	16.5±0.9	16.5±1	16.6±1.3	14.5	16.1±0.1	15.9±1.3	16±1.1	16±1.5	15.9±0.8	16.6±2.2	18.1±4.1
pH	8.2±0.4	8.3±0.3	8.4±0.3	8.4±0.2	8.2±0.3	8.4±0.4	8.8	8.6±0.3	8.7±0.2	8.6±0.1	8.6±0.04	8.6±0.3	7.8±1.1	8.9±0.1
Eh	391.6±115.9	329±143	234.5±93	164.4±145.1	175.6±94.9	194.9±210.2	302.5	374±7	381.4±37.4	392.7±31.9	247.1±60.8	353.4±66.7	331.5±65.5	277.6±48.3
EC	162±0.9	133.6±1.6	145.8±3.7	161.9±18.9	180.5±0.8	174.6±1.3	200.1	139.8±16.7	131.8±4.7	133.2±12.9	118.4±2.3	136.7±1.1	135.7±5.9	123.5±1.6
DO	0.05±0.03	0.97±0.4	0.04±0.01	0.02±0.01	0.04±0.01	0.03±0.01	0.38	1.3±1.1	0.9±0.5	1.3±0.9	1.6±0.5	2.8±1.2	0.9±0.4	1.2±0.3
Na	10.8±0.4	12.6±0.3	17.4±3.2	18.5±2.8	20.0±0.5	23.1±1.9	41.5±0.5	13.8±0.5	12.9±0.2	11±0.9	12.3±0.1	12.7±0.9	10.8±1.3	17.2±0.1
K	0.5±0.1	0.2±0.01	0.4±0.2	0.5±0.4	0.4±0.1	0.3±0.1	0.5±0.3	0.6±0.7	0.4±0.3	0.4±0.3	1.3±0.2	1.1±0.3	1.6±0.3	0.4±0.4
Ca	19.5±1	15±0.4	16.2±3.1	12.7±3.1	15.4±1.4	13.4±1.5	7.3±1	18.5±0.7	16.4±1.1	16.4±1.4	13.4±0.8	16.2±1.5	18.3±0.5	10.5±1.2
Mg	2.4±0.2	0.4±0.02	1.1±0.1	1.5±0.3	2.3±0.1	1.2±0.2	0.4±0.02	1.2±0.5	0.4±0.03	0.4±0.03	0.5±0.1	0.6±0.02	0.4±0.1	0.2±0.03
SiO_2	17.6±1.7	16.8±1.4	19.4±3.3	17.3±2.7	19.3±1.9	18.3±1.8	17.3	16.2±1.4	15.5±1.3	19.1±1.4	7.2±1.3	11.8±1.3	17.8±2.5	15.2±1.1
Cl	2.1±0.1	2.5±0.2	2.5±0.4	1.9±0.3	1.9±0.1	1.8±0.1	1.9±0.2	2.9±0.1	2.6±0.1	2.6±0.1	2.7±0.1	3.1±0.1	3±0.3	2.3±0.1
SO4	6.4±0.2	6.0±0.1	5.9±0.4	5.8±1.2	5.3±0.04	6.1±0.1	2.7±1.4	9.2±0.6	6.9±0.4	4.2±0.3	2±0.5	4.5±0.8	6.9±0.4	6.9±0.5
F	4.0±0.2	4.4±0.4	5.4±0.4	4.7±0.6	5.2±0.4	6±0.7	9.3±0.8	4.7±0.8	4.6±0.1	3.7±0.1	3.1±0.1	3.8±0.3	3.6±0.3	6.5±0.2
HCO_3	78.1±5.1	60.7±2.4	69.8±2.4	68.9±11.7	91.4±6	88.5±9.3	97.7±4.2	63.1±1.8	60±1.8	58±3.1	56.9±1.8	61±5.3	58±5.3	58±0
Mn	0.7±0.3	0.2±0.2	21.2±5.1	18±12.9	3.6±0.7	2.7±0.7	6.3±2.1	10.3±1.4	1.9±2.1	0.2±0.1	10.3±0.3	46.6±4.6	11.2±1.8	4±0.3
U	40.1±2.5	672.1±65.4	26.6±12.8	10.1±4.5	6.7±2.5	4.7±0.8	1.1±0.3	575.8±175.9	878±44.2	142.7±8.4	10.3±2.2	189.3±66.8	583.7±69.1	25.6±7.2
$\delta^{18}O$	-8.8±.0.1	-8.5±0.1	-8.8±0.1	-8.7±0.1	-8.7±0.2	-8.6±0.3	-8.8±0.3	-8.7±0.1	-8.6±0.1	-8.6±0.1	-8.8±0.1	-8.6±0.1	-8.6±0.1	-8.9±0.1
δD	-61.9±0.8	-60±0.6	-61.7±0.5	-61.5±0.8	-62.8±0.8	-61.6±1.5	-64.8±1	-59.8±0.6	-59.5±1.2	-59.3±0.3	-59.9±0.2	-58.7±1	-59.2±0.5	-62.1±0.3

Uranium speciation in granitic groundwater and isotopic study

U speciation (I)

❑ Calculation of U species

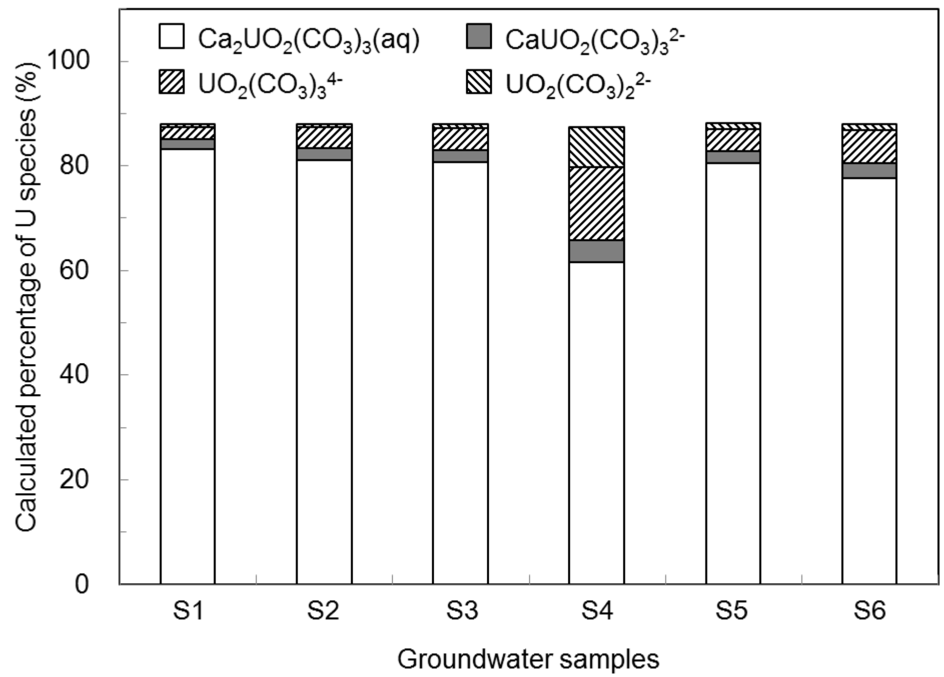
- GWB (Geochemist's Workbench, Professional Version 9.07)
- Thermochemical Database: NEA-TDB Data + **(Ca-UO_2-CO_3)** data (Bernhard et al., Radiochim Acta, 2001)

  - For $CaUO_2(CO_3)_3^{2-}$, log K° = 25.40

  - For $Ca_2UO_2(CO_3)_3(aq)$, log K° = 30.55
- Dominant U species : **$Ca_2UO_2(CO_3)_3(aq)$**
- Similar distribution of U in all GW samples
- Major controlling factor: **$[Ca^{2+}]$**

  → When $[Ca^{2+}]$ is low (S4),

  the percentages of $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ increase



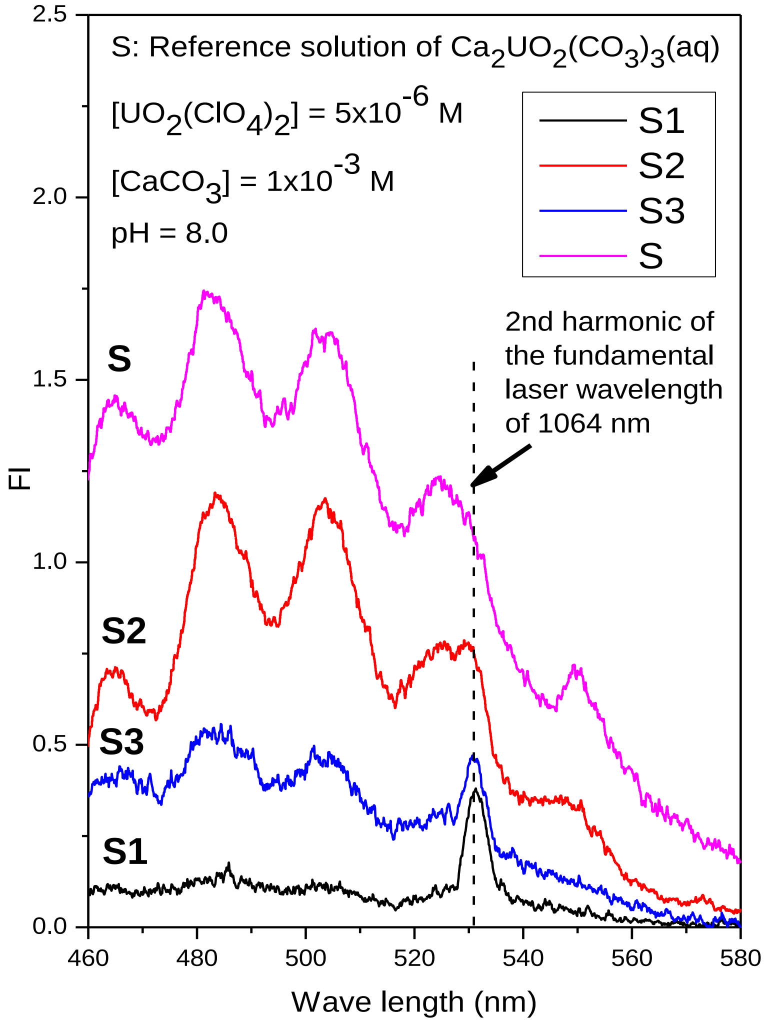
"GWs sampled from DB-1"

Baik et al., JRNC 305, 2015

U speciation (II)

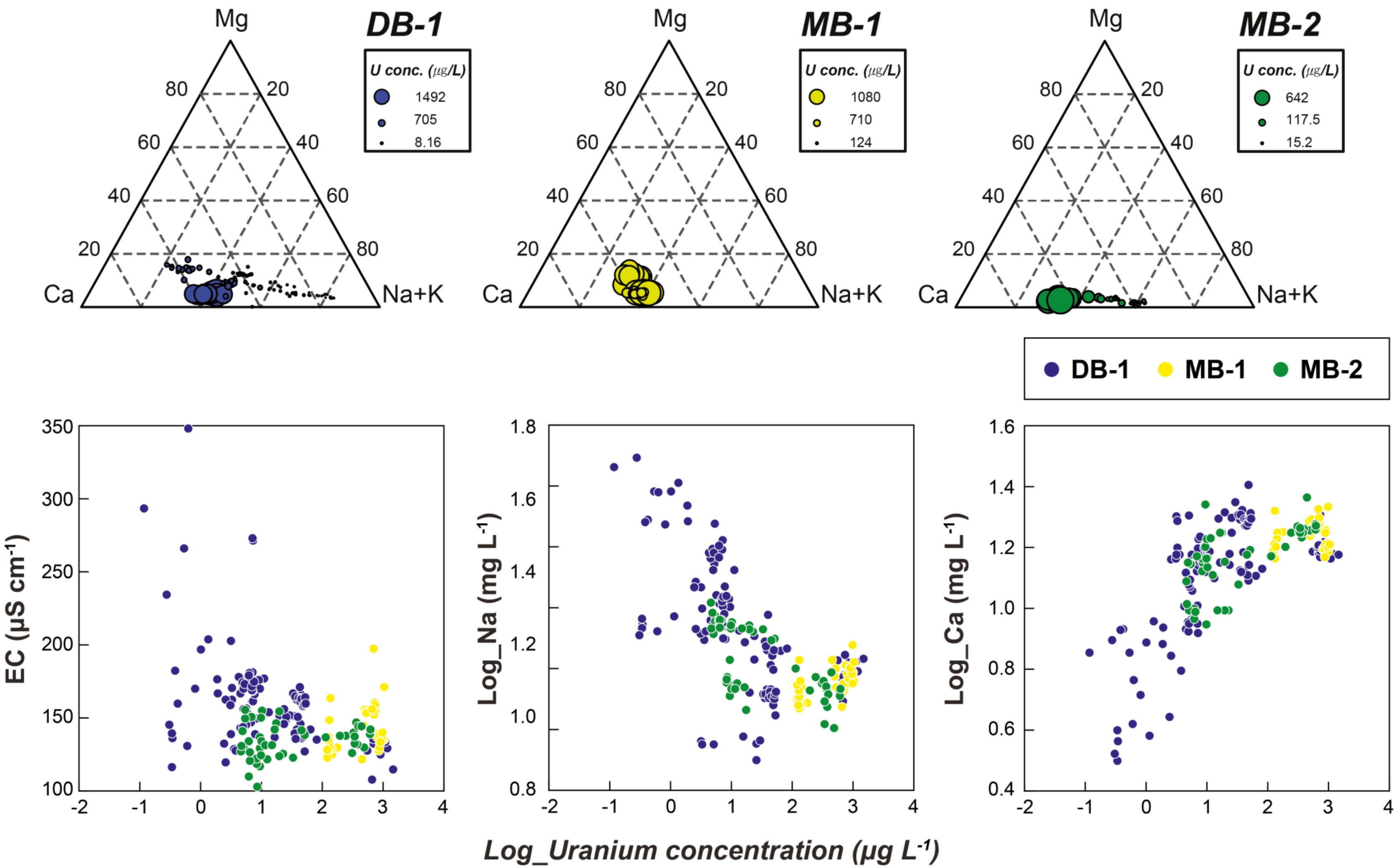
❑ Identification of U species by TRLFS

- Gate delay: 40 ns
- Gate width: 100 ns
- In some groundwater samples (S1, S2, and S3),
  - → Peak positions: 465, 484, 504, and 524 nm
  - → FL life time = 37 ± 3 ns
  - → Dominant U species is **$Ca_2UO_2(CO_3)_3(aq)$**
  - → Agree with the result of model calculation
- When U was low (< 50 μg/L),
  - → Difficult to identity the U species
  - → Mainly due to organic materials



Baik et al., JRNC 305, 2015

U vs. Ca



✓ ***High dissolved U in groundwater exists under Ca dominant environment***

U behaviors

❑ AR of $^{234}U/^{238}U$ with Depths in KURT Groundwater

- At shallower depths of an oxidized condition, strong leaching of U occurs
  → High U content and AR near to 1.0
- At deeper depths of a reduced condition, dissolution of ^{234}U by α-recoil
  → Low U content and AR > 1.0

❑ Anomaly of high U content

- U^{tot} > 100 μg/L in KAERI groundwater
- The study site has not known to contain U orebody or mineralization

❑ Cause of the enhanced U in the granitic KURT groundwater

- High uranium content in rock
- High concentration of Ca
- Strongly oxidizing condition

Summary

- **In this study, the distribution and potential migration of natural uranium as well as uranium species were evaluated to investigate geochemical behavior of uranium in fractured granitic bedrock.**

- ❑ **In Korea, the groundwater wells containing high natural uranium located predominantly in the plutonic bedrock region.**

- ❑ **KAERI groundwater samples from specific regions in the boreholes monitored showed an anomaly in U concentration, ranged of ~ mg/L in units.**

- ❑ **Water conductive feature as a migration pathway of uranium were traced during the KURT extension.**

- ❑ **From TRLFS measurement and model calculation, uranium mainly exists as $Ca_2UO_2(CO_3)_3(aq)$ ternary complex in the KURT groundwater**

- ❑ **Natural uranium can be used as an analogue for the geochemical behavior of radionuclides in HLW disposal environments, which is very important in the safety case for the deep geological disposal of HLW.**

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