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Radioactivity Concentrations of ^{3}H and ^{137}Cs in Coastal Groundwater in Korea

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Abstract

The activities of tritium (^{3}H) and cesium-137 (^{137}Cs) were measured in coastal brine groundwater and groundwater in the Republic of Korea to establish baseline data relevant to evaluating potential effects associated with the discharge of ALPS-treated water from the Fukushima Daiichi Nuclear Power Station. Tritium activities in the brine groundwater samples ranged from 1.29 to 9.05 TU (0.15–1.07 Bq/L), whereas ^{137}Cs activities were below the sample-specific detection limits (1.82–2.86 mBq/L). The low ^{137}Cs activities are consistent with the potential retention of cesium by soil and rock during seawater infiltration; however, the present data do not independently quantify the extent of this retention. These measurements provide baseline data for future monitoring.

1. Introduction

Following the Great East Japan Earthquake in 2011, the Fukushima Daiichi Nuclear Power Station accident resulted in the loss of reactor cooling functions. Large volumes of contaminated water, generated in part by the inflow of groundwater and seawater, were treated using the Advanced Liquid Processing System (ALPS) and stored on site. Since 2023, the Japanese government has conducted multiple controlled discharges of ALPS-treated water into the Pacific Ocean. Approximately 4.5 trillion Bq of tritium were released in 2023 and 5.7 trillion Bq in 2024 [1,2]. The Japanese government has stated that the treated water meets the applicable discharge criteria, and the International Atomic Energy Agency (IAEA) has reviewed relevant safety aspects and monitoring arrangements [3]. Public and scientific discussion has nevertheless continued regarding potential effects on marine ecosystems, seafood, and coastal environments [4–7].

Radioactive cesium has continued to be detected in some food products in Japan more than a decade after the Fukushima Daiichi accident. According to the Japanese Ministry of Health, Labour and Welfare, radioactive cesium was detected in 9.4% of tested agricultural, livestock, and fishery products in 2024, with particularly high detection rates in wild meat and agricultural products.

In the Republic of Korea, environmental radioactivity monitoring has been conducted since the

Fukushima Daiichi accident. National and regional monitoring programs cover marine waters, coastal and port areas, marine organisms, and seabed sediments [8–10]. Saline groundwater is also used for land-based aquaculture in coastal areas, including Jeju Island. Such water can reflect mixing between seawater and fresh groundwater and interactions with subsurface materials. Establishing baseline radionuclide concentrations in these aquifers is therefore useful for future environmental monitoring. In this study, tritium (^{3}H) and cesium-137 (^{137}Cs) were measured in coastal groundwater and brine groundwater collected from sites on Jeju Island and along the East, South, and West coasts of Korea beginning in June 2024. Low-level analytical methods, including electrolytic enrichment for tritium and chemical concentration for cesium, were used.

2. Experimental Methods

2.1. Sampling and hydrochemical analysis

For brine groundwater, 60 L samples were collected for ^{137}Cs analysis and 1 L samples for ^{3}H analysis using submersible pumps after in situ water-quality measurements. Sampling locations are listed in Table 1 and shown in Figures 1. Field parameters—pH, electrical conductivity (EC), oxidation–reduction potential (ORP), dissolved oxygen (DO), and temperature—were measured using a portable multiparameter meter (WTW Multi 3620 IDS; instrument manufacturer and country should be verified). The stated uncertainty of EC measurements was ±0.5%. Major cations (Ca, Mg, Na, and K) and anions (F, Cl, Br, NO_3^-, and SO_4^{2-}) were analyzed in the laboratory and the analytical results were shown in Table 2. Samples were filtered through 0.45-μm cellulose acetate membranes, stored in polypropylene bottles, and refrigerated at 4 °C before analysis. Samples for cation analysis were acidified to pH < 2 with ultrapure nitric acid. Cation and anion concentrations were determined by inductively coupled plasma optical emission spectrometry (ICP–OES; Optima 7300 DV, PerkinElmer, USA) and ion chromatography (Dionex Aquion; Thermo Scientific), respectively. Charge-balance errors were within ±10%.

2.2. Tritium and ^{137}Cs analysis

For ^{3}H analysis, 1-L samples were distilled and concentrated approximately 20-fold using an electrolytic-enrichment system, and the enriched samples were measured by liquid scintillation counting (LSC) [11]. For ^{137}Cs analysis, 60-L samples were acidified to approximately pH 1.5 with hydrochloric acid, and ^{134}Cs was added as a yield tracer to estimate chemical recovery. Cesium was coprecipitated with ammonium phosphomolybdate (AMP), and the precipitate was separated, redissolved in 10 M NaOH, and analyzed by gamma spectrometry [12,13].

3. Results and Discussion

Long-term monitoring of marine radioactivity around Korea has been conducted since 2018. According to the cited monitoring report, ^{137}Cs concentrations in surface seawater at 40 stations in 2023 ranged from 0.794 to 1.93 mBq/kg, with an annual mean of 1.29 ± 0.20 mBq/kg. For 2018–2022, the reported range was 0.797–2.26 mBq/kg, with a mean of 1.45 ± 0.23 mBq/kg; values for 2006–2010 were reported as <1.19–4.04 mBq/kg [14]. Tritium concentrations in surface seawater in 2023 were reported as 0.0872–0.448 Bq/L, with a mean of 0.187 ± 0.072 Bq/L. These analytical results were comparable to previous five years (2018-2022) data ranged from 0.0577 to 0.451 mBq/kg(mean: 0.173±0.068 Bq/L) and these values were lower than pre-accident levels. This radionuclide activity survey was only monitored sea water and coastal area effect was not surveyed. To evaluate radionuclide behavior in coastal aquifers, groundwater and brine groundwater samples were analyzed and compared with seawater. Groundwater and brine groundwater from the KR, KRP, and JJ areas were therefore compared using the data in Tables 3–5. Tritium results vary among individual samples and sites. At KR, most groundwater samples were below the reported quantification threshold, whereas some stream or groundwater samples had measurable tritium. At KRP, tritium values also varied between samples. The suggestion that relatively high tritium in the KRP seawater sample reflects mixing with surface water because EC value at KRP site was low rather than other site, therefore, ^{3}H concentration was high due to high surface water ^{3}H concentration. The ^{3}H concentration at JJ site was also showed groundwater mixing effect. JJ sample was sample with depth, so ^{3}H concentration was decreased with depth and EC value increasing.

Brine groundwater samples from the East Sea (ESG), South Sea (SSG), West Sea (WSG), and Jeju Island (JSG) were analyzed for ^{3}H and ^{137}Cs; the results are presented in Table 6. The EC values suggest differing degrees of mixing between seawater and fresher groundwater. Several Jeju samples (JSG-1, JSG-3, and JSG-4) have EC values close to that of the seawater reference, whereas samples with lower EC may contain a larger freshwater contribution. This interpretation should be supported by the major-ion data and clearly defined criteria for classifying samples. The elemental relationships showed in Fig. 2, in the coastal brine groundwater samples indicate that chloride concentrations generally increase with electrical conductivity (EC), sodium (Na), sulfate (SO_4), and bromide (Br). This pattern is consistent with the influence of seawater on the hydrochemical composition of coastal brine groundwater. The Jeju samples JSG-1, JSG-3, and JSG-4 exhibit EC and major-ion concentrations close to those of the seawater reference, suggesting a substantial marine contribution. In contrast, several samples from the East and West Sea regions exhibit lower EC and chloride concentrations, indicating potentially greater contributions from freshwater groundwater. Nevertheless, these relationships alone cannot quantify the mixing proportions or distinguish seawater mixing from other geochemical processes, such as mineral–

water interactions and ion exchange. Further evaluation using major-ion ratios, stable isotopes, and appropriate mixing models would help clarify the processes controlling the observed hydrochemical variability. The Piper diagram(Fig. 4) indicates predominantly Na–Cl-type water, consistent with a substantial marine contribution. The ^{3}H content of most brine groundwaters was less than 2 TU (0.2 Bq/L), and when the groundwater samples were mixed with brine groundwater, the ^{3}H content was high. In the case of Jeju region, in the case of samples collected by depth of well (JJ-1~5), the deeper the depth, the closer it was to seawater, and the ^{3}H content decreased.

The mean chemical recovery for the AMP coprecipitation procedure applied to 60-L samples was reported as 71 ± 8%. All ^{137}Cs results in Table 6 are below their respective sample-specific detection limits, which range from 1.82 to 2.86 mBq/L. These results are consistent with low ^{137}Cs activity in the analyzed samples. These low activity was occurred by sorption or retention by soil and bedrock during seawater infiltration[15,16].

4. Conclusion

This study measured ^{3}H and ^{137}Cs in coastal brine groundwater in Korea to establish baseline data relevant to monitoring coastal aquaculture water. The ^{3}H results reported in Table 6 range from 1.29 to 9.05 TU, while ^{137}Cs activities were below the sample-specific detection limits of approximately 1.82–2.86 mBq/L. The tritium data should be reported consistently in TU and/or Bq/L, with the conversion factor stated. Because the measured tritium values do not all fall below 0.2 Bq/L, any broader statement that all or most samples were below this level should be revised to match the data. The ^{137}Cs results provide a baseline for future comparisons, but the data alone do not prove that cesium was removed by sorption in surrounding rocks and soils. Repeated sampling and consistent analytical quality assurance would strengthen future assessments of temporal trends.

Acknowledgements

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Conflicts of Interest

The authors declare no conflict of interest.

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Table 1. Brine groundwater sampling location and field water quality data

sample	longitude	latitude	Temp. (°C)	pH	EC (μS/cm)	DO (mg/L)	ORP (mV)
WSG-1	36°8' 15.19"N	126°34'42.19"E	16.5	7.35	12,190	0.08	-18.3
WSG-2	35°38' 1.51"N	126°28'56.13"E	15.3	6.93	5680	0.287	93.1
WSG-3	35°3' 3.25"N	126°14'9.90"E	16.5	7.27	2610	0.394	-176.8
ESG-1	38°20' 6.69"N	128°31'18.29"E	11.9	6.84	5,980	0.657	-81.6
ESG-2	37°37' 59.73"N	129° 2'45.54"E	14.8	9.80	576	1.81	-405.6
JSG-1	33°32' 31"N	126° 24' 45"E	16.8	7.83	49,300	6.45	65.2
JSG-2	33°25' 31"N	126° 54' 52"E	17.4	7.83	29,000	8.53	89.1
JSG-3	33°33' 23"N	126° 49' 23"E	17.8	8.21	48,600	9.7	82
JSG-4	33°18' 30"N	126° 49' 48"E	18.5	8.20	49,700	6.91	35.4
JSG-5	33°14' 39.79"N	126°12'42.01"E	17.5	7.07	10,100	8.12	76.3
SSG-1	34°40' 10.68"N	127° 5'11.44"E	19.5	7.14	30,300	2.05	117.7
SSG-2	34°54' 16.97"N	128°27'28.65"E	17.2	6.99	1,118	6.76	218
KR	37°46' 2.64"N	128°56' 29.72"E	-	-	-	-	-
KRP	35°59' 10.50"N	129°33' 15.57"E	-	-	48,370	-	-

Table 2. Results of trace element content in brine groundwater

Sample	EC (μS/cm)	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	SiO_2 (mg/L)	Sr (mg/L)	B (mg/L)	Li (mg/L)	HCO_3 (mg/L)	F (mg/L)	Cl (mg/L)	Br (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
WSG-1	12190	238	262	1950	68.8	13.1	2.18	<0.3	<0.1	740	<1	3770	12.1	11.9	<1	481
WSG-2	5680	230	82.1	786	8.21	26.2	3.57	<0.3	0.494	130	<1	1610	5.45	15.6	<1	266
WSG-3	2610	122	53.6	55.1	4.03	40.8	1.22	<0.03	<0.01	98.7	0.111	398	1.46	0.4	<0.1	20.3
ESG-1	5980	37.2	100	904	3.92	3.88	0.517	<0.3	<0.1	37.7	<1	1680	5.44	<1	<1	246
ESG-2	576	25.3	11.7	51.4	1.62	0.364	0.061	<0.03	<0.01	16.4	<0.1	101	0.224	<0.1	<0.1	87.2
JSG-1	49300	356	1160	10200	333	18.7	6.5	3.95	<0.1	134	<2	18400	61	<2	<2	2490
JSG-2	29000	202	642	5690	188	19.4	3.64	2.32	<0.1	97.3	<2	10100	35.3	7.99	<2	1370
JSG-3	48600	352	1130	10100	341	4.56	6.6	4.03	<0.1	134	<2	17800	59.7	<2	<2	2450
JSG-4	49700	349	1130	10100	340	3.96	6.46	3.99	<0.1	142	<2	17900	59.7	<2	<2	2450
SSG-1	30300	216	635	5770	193	12.6	3.94	251	<0.1	123	<2	10300	33.7	10	<2	1370
SSG-2	1118	46	15.7	139	2.2	22.5	0.376	<0.03	<0.1	120	<0.1	252	0.854	24.3	<0.1	41.8
Sea water	51450	410	1,350	10,500	390	6.4	8	4.5	0.17	0.00	1.3	19,000	67			2,700

Table 3. Isotope and H-3 content in groundwater and brine groundwater in KR area

Sample	EC (μS/cm)	$\delta^{18}O$ (‰)	δD (‰)	^{3}H(TU)	
KR-1	23,760	-4	-31.6	3.0±0.3	brine
KR-2	1,373	-8.2	-53.2	<0.3	groundwater
KR-3	6,891	-7.2	-51.6	<0.3	brine
KR-4	854	-6.8	-50.4	<0.3	groundwater
KR-5	524	-8.8	-62.7	<0.3	groundwater
KR-6	1,604	-7.9	-54.5	4.4±0.4	stream
KR-7	1,348	-8.4	-54	<0.3	groundwater
KR-8	174	-7.8	-55.8	2.9±0.2	groundwater
KR-9	-	-0.2	-1.5	<0.3	sea water

Table 4. Isotope and H-3 content in groundwater and brine groundwater in KRP area

Sample	EC (μS/cm)	$\delta^{18}O$ (‰))	δD (‰)	^{3}H(TU)	
KRP-1	48,370	-0.7	-10.2	<0.3	brine
KRP-2	3,621	-6.7	-49.4	2.2±0.2	groundwater
KRP-3	2,062	-9.2	-61.8	<0.3	groundwater
KRP-4	205	-8.5	-56.7	2.0±0.2	groundwater
KRP-5	10,730	-6.5	-47.7	1.9±0.2	brine
KRP-6	1,705	-5.7	-42.0	3.7±0.4	groundwater
KRP-7	2,492	-7.1	-52.7	0.7±0.1	groundwater
KRP-8	26,450	-3.3	-22.2	4.1±0.3	sea water
KRP-10	32,240	-2.2	-20.1	<0.3	brine

Table 5. Isotope and H-3 content in groundwater and brine groundwater in JJ area

Sample	depth (m)	EC (μS/cm)	^{3}H(TU)	
JJ-1-1	15	12,060	5.53±0.35	brine
JJ-1-2	25	31,621	2.84±0.21	brine
JJ-1-3	35	44,666	2.58±0.29	brine
JJ-1-4	60	44,016	1.82±0.09	brine
JJ-2-1	25	2,885	2.53±0.21	brine
JJ-2-2	50	33,261	2.81±0.21	brine
JJ-2-3	65	41,261	1.93±0.22	brine
JJ-2-4	80	41,111	1.49±0.13	brine
JJ-3-1	50	626	3.06±0.23	brine
JJ-3-2	80	37,529	1.59±0.14	brine
JJ-4-1	50	680	2.53±0.22	brine
JJ-4-2	80	36,710	1.18±0.09	brine
JJ-5	-	41,261	1.90±0.15	brine
JJ-6	-	239	2.57±0.18	groundwater
JJ-7	-	288	4.02±0.39	groundwater
JJ-8	-	87	3.27±0.23	groundwater
JJ-9	-	45,752	1.45±0.13	sea water

Table 6. Results of concentration of radioisotopes in brine groundwater

sample	^{3}H(TU)	Cs-137(mBq/L)
ESG-1	5.15±0.30	<2.03
ESG-2	8.71±0.64	<2.22
SSG-1	2.53±0.26	<1.83
SSG-2	4.45±0.39	<1.82
WSG-1	9.05±0.91	<1.84
WSG-2	1.48±0.15	<2.00
WSG-3	4.34±0.34	<1.94
JSG-1	1.29±0.11	<2.86
JSG-2	2.36±0.23	<1.99
JSG-3	1.43±0.13	<1.85
JSG-4	1.72±0.09	<1.97
JSG-5	2.97±0.27	<1.99



Fig. 1. Brine groundwater sampling site

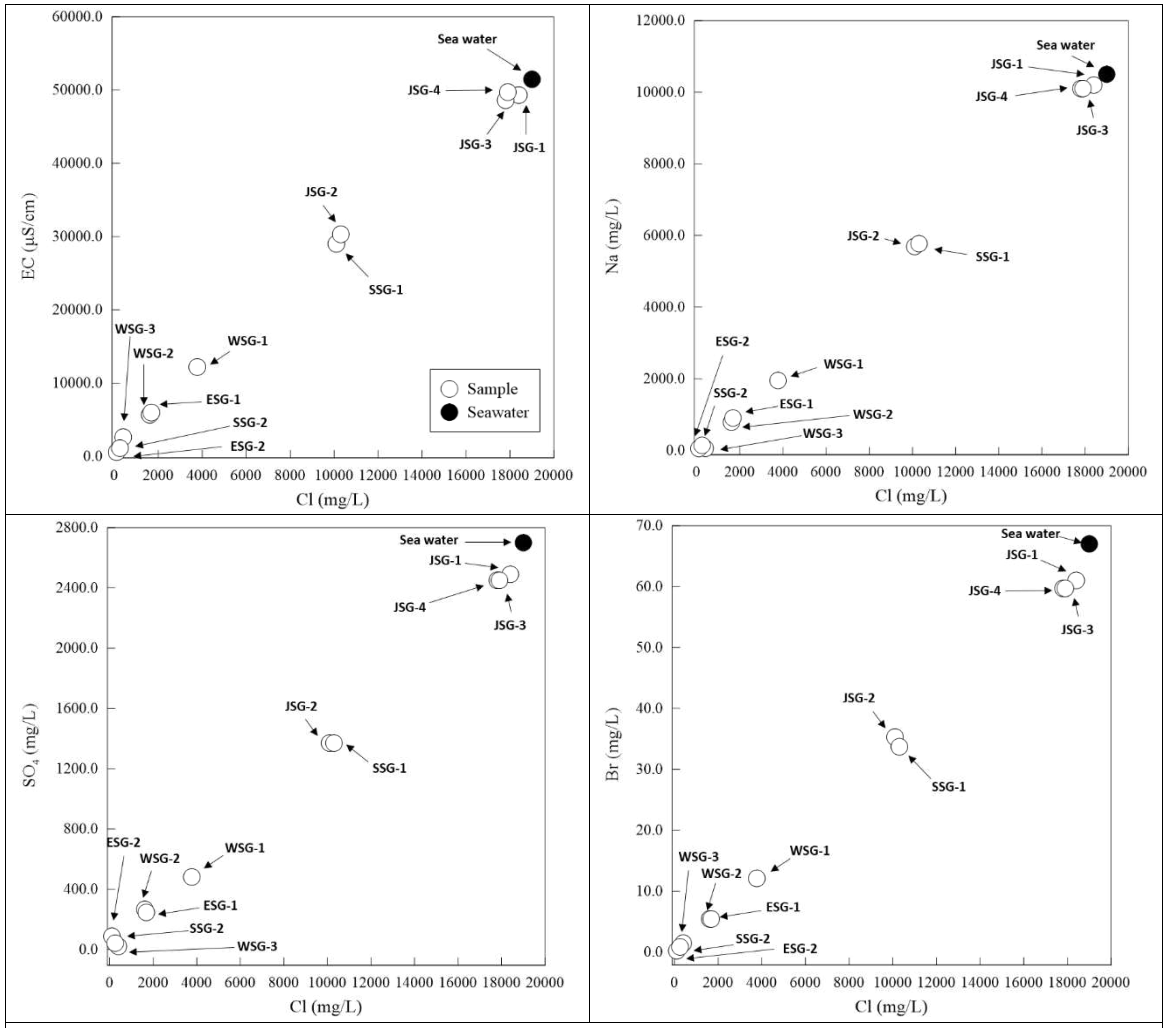


Fig. 2. Elemental correlations in brine groundwater samples

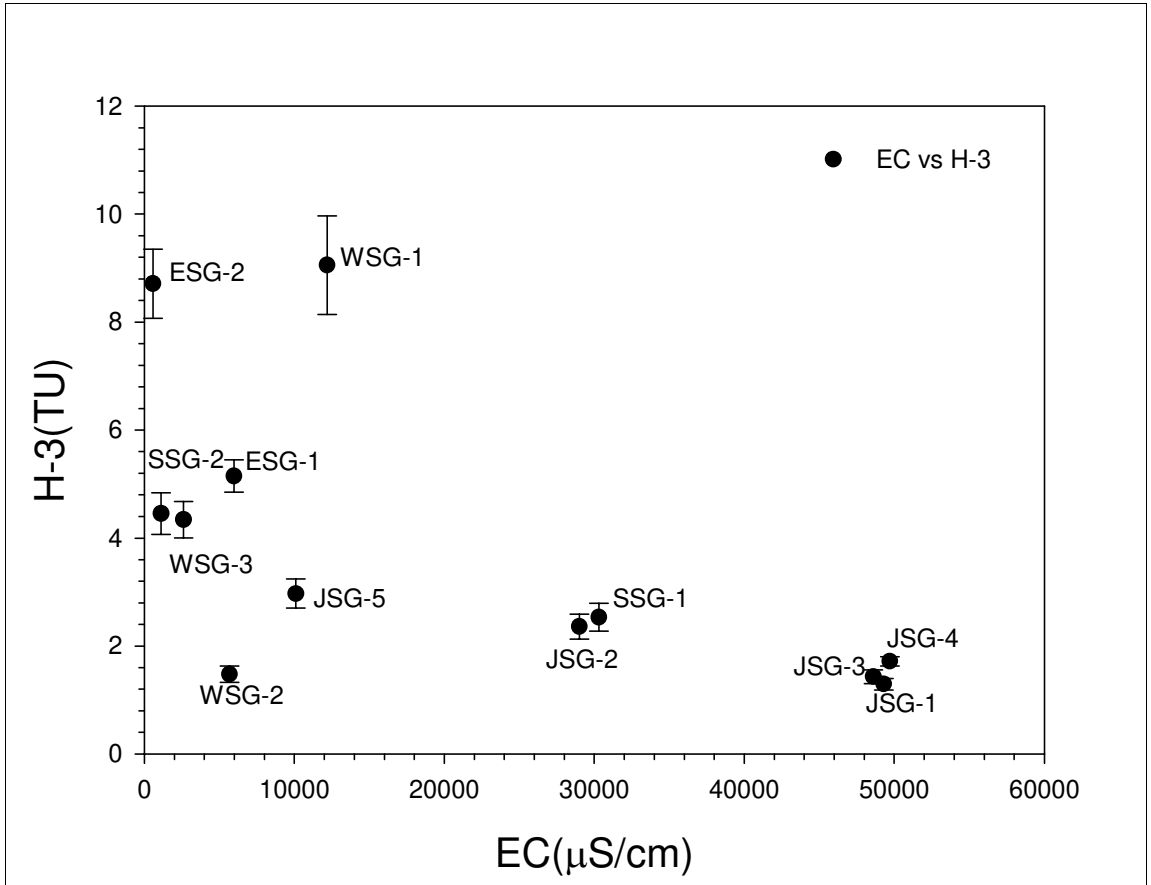


Fig. 3. EC vs. H-3 content relationship in brine groundwater samples

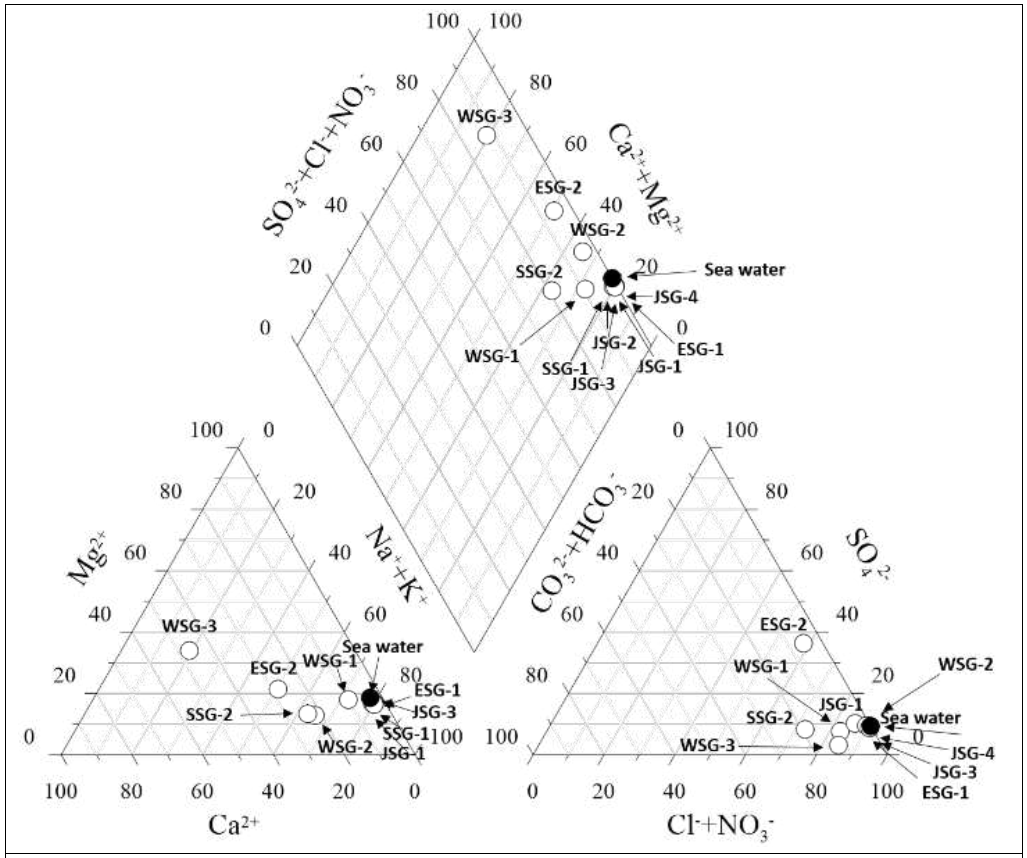


Fig. 4. Piper diagram of brine groundwater sample.