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Stable Isotopes and Hydrochemistry Approach for Determining the Salinization Pattern of Shallow Groundwater in Alluvium Deposit Semarang, Central Java

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Abstract - A groundwater study has been conducted in the Semarang City in August 2014, aiming to determine the source of shallow groundwater salinization using stable isotopes (^{18}O, ^{2}H) and water chemistry approach, and supported by local hydrogeological data. A number of shallow groundwater samples were taken at several locations with a depth of 0 - 35 m. Based on geological data, shallow groundwater of Semarang alluvium is dominated by insertion of sand-gravel and sandy-clay with average porosity of around 56.0 %. This layer is thinning towards the south and then increasingly thickening to the north and north-east of the studied area. The results of the analyses show that the characteristics of shallow groundwater, i.e. approximately 51% of groundwater, still have the original composition as meteoric water and the remaining approximately 49% obtained a shift in the isotopic composition as caused by interaction with seawater and the little influence of evaporation. The results of chemical analysis of water indicates that in dry seasons, shallow groundwater aquifers in the Semarang City is dominated by chloride (Cl-) with NaCl of water type. While the parameters of bicarbonate, chloride, and Na/Cl ratio, shallow groundwater can be classified into two groups, namely unintruded groundwater around 51% spread from the foot hills to the south towards the hills with elevations of 9 - 142 m above sea level and intruded groundwater around 49% spread from the coastline to the urban direction.

Keywords: stable isotopes, hydrochemistry, salinization, shallow groundwater, alluvium, Semarang

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Introduction

Semarang is a developing city that evolved from an old residential place built above alluvial deposits. Recently, Semarang grows rapidly and becomes a province capital city and one of important cities in Indonesia (Wardhana *et al.*, 2014). The industrial growth as well as business and trading growth in Semarang have influenced the need of clean water, which mainly depends on groundwater supply (Irham *et al.*, 2006; Katrinavia *et al.*, 2015). Most of residents in Semarang plain utilizes shallow groundwater taken from 3 - 18 m below the ground level, while the residents in higher elevation utilize shallow groundwater taken from 20 - 40 m below ground level (BPS Kota Semarang, 2009). Due to high rate groundwater extraction, Semarang City experiences land

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subsidence up to 19 cm/year (Abidin *et al*., 2012) that leads to extensive coastal flooding (rob, in local language).

Another effect of excessive groundwater extraction is seawater intrusion (Mas-Pla *et al*., 2013) which leads to groundwater salinization. In recent years, increasing phenomena of groundwater salinization were found in several locations in Semarang area, mainly in residential areas and several industrial areas in the northern part, *i.e*. North Semarang, Central Semarang, and Gayamsari. This increasing salinization is monitored from several shallow dug wells and deep wells scattered around the city. Recently, a salinization phenomenon is estimated to reach 4 km inland from the coastline (Suhartono *et al*., 2015).

Salinity is commonly defined as the weight of inorganic ions (gram) dissolved in 1 kg of water (Gat, 2010). Salinization is one of several factors that lead to the decrease of groundwater quality. This process can be identified using several techniques such as water chemistry and isotopic tracer of studied groundwater (Han *et al*., 2011; Gassama *et al*., 2012). Water chemistry can be used to classify groundwater type and distribution of salinization using Piper diagram and chloride-bicarbonate ratio (Chekirbane *et al*., 2013; Wen *et al*., 2012). While water stable isotope content (^{18}O and ^{2}H) can be used to infer the origin and genesis of groundwater (Mook, 2000).

Hydrogeology of Semarang

According to Sudaryanto and Wibawa (2003), the landscape of Semarang City consists of coastal plain and hilly area between 0 - 270 m above sea level (Figure 1). The morphology of coastal plain area is about 0 - 50 m above sea level, spreading from Kendal coastal area in the west, and Semarang and Demak in the east. This morphology is covered with alluvium deposit consisting of river deposit, Garang delta deposit, and coastal deposit (Sudaryanto and Wibawa, 2013). Generally, the coastal area is composed of clays and sands with typical thickness of about 50 m. While river and lake deposits are composed of gravels, pebbles, sands, and silts with typical thickness of about 1 - 3 m (Mamlucky and Dadi, 2007).

Based on geology and its formation, the groundwater system in Semarang City area is divided into two groundwater systems as follow (Marsudi, 2001):

Lower Plain Aquifer

This aquifer consists of an unconfined aquifer with the depth of 5 - 30 m below ground level and a confined aquifer composed of Delta Garang and Damar Formation aquifers. The depths of both aquifers are about 30 - 130 m below ground level and semiconfined to confined in nature. The lithology of the main aquifer in Delta Garang Formation mainly consists of medium- to coarse-

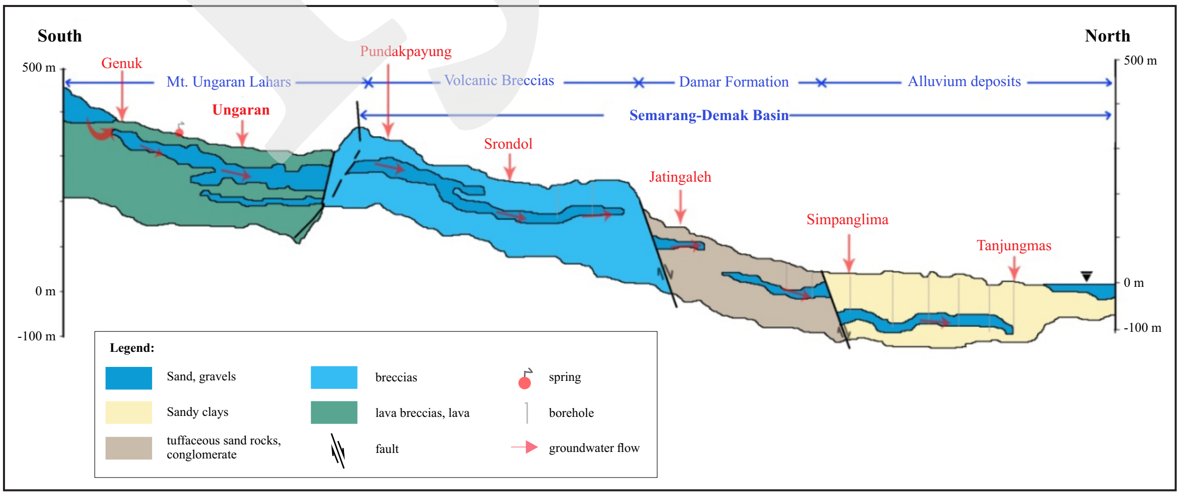

Figure 1. Hydrogeological cross-section of Ungaran-Semarang area (Taufiq, 2010).

grained sandstone or conglomerates. While main aquifer of Damar Formation comprises conglomerates and tuffaceous sandstone overlain by an impermeable layer of clay, tuff, and breccias (Mamlucky and Dadi, 2007).

Aquifer in Hilly Area

There are two aquifers in the hilly area of Semarang. The first one is unconfined aquifer, *i.e.* located at Notopuro Formation and Old Ungaran Formation. The water table in these formations is about 8 - 15 m below the ground level with the thickness of around 10 - 30 m. However at some locations, the water table could reach 20 m depth, while in other locations the groundwater emerges at surface as springs. The second is confined aquifer which consists of volcanic sands, volcanic breccias, and conglomerates form Damar Formation that spread across the hilly area. This aquifer is normally found between 30 - 150 m below the ground level.

Sriyono *et al.* (2010) conducted a hydrostratigraphy study using the geo-electrical method to find out the stratification of rock and aquifer-forming materials. The study was done in seven locations in Semarang, *i.e.* Tanah Mas, Tawang Mas, Masjid Agung, East Semarang, Pekunden, Perum Penerbad, and Polsek Sembar. The similar study was also done by Sophian (2002) using three deep boreholes in Semarang, *i.e.* Tawang Sari, Distamben, and Poncol Stations. Based on those studies, the shallow aquifer of Semarang City consists of sand-pebbles and sand-clays with the average porosity of 55.95% and situated 0.5 - 16 m below the ground level.

Materials and Methods

Stable Isotope

Stable isotopes of ^{18}O and ^{2}H form water molecules in form of $^{1}H_2{}^{18}O$ and $^{1}H_2H^{16}O$ (Gibson and Reid, 2010). These molecules are sensitive to physical processes such as evaporation and condensation and can undergo fractionation that can alter the isotopic ratio (Allen *et al.*, 2013). Therefore, to prevent fractionation, necessary precautions were taken while taking water samples, *i.e* sample bottle must be as close as possible to the water discharge or submerged below water surface in case of spring samples. All water samples were contained in 20 ml bottle without air bubbles. The analysis of stable water isotopes was conducted in Hydrology Lab, PAIR - BATAN, using liquid-water stable isotope analyzer LGR (Los Gatos Research) DLT-100.

Standard mean oceanic water (SMOW) was arbitrarily defined to have isotopic content of 0‰ for both ^{18}O and ^{2}H, while meteoric water has more depleted isotopic content than the seawater due to fractionation caused by physical processes. Thus, groundwater that already mixed with seawater due to intrusion was expected to have more enriched isotopic content compared to normal groundwater and plotted along a mixing line on graph of $\delta^{18}O$ *vs.* $\delta^{2}H$ (Figure 2). This graph was a common tool to identify seawater intrusion on coastal areas (Swarzenski *et al.*, 2013).

Hydrochemistry

The amount of water samples needed for chemical analysis was 1L and than analyzed using ion chromatograph for major ion composition *i.e.* Cl-, SO_4^{2-}, Na^+, Ca^+, Mg^+ and K^+. While HCO_{3-} was analyzed using the titrimetric method. These ions can be useful to identify the type and chemical evolution of waters which can be visualized using a Piper diagram which gives a quick view on water classification and evolution, *e.g.* normal groundwater was plotted around HCO_3 apex, seawater is plotted in Na-Cl corner, thus seawater-contaminated groundwater is plotted towards Na-Cl corner.

In combination, the distribution pattern of seawater intrusion as well as contamination level in the studied area was approached by using stable isotopes and hydrochemistry method which was deployed by using the graph of $\delta^{18}O$ *vs.* chloride ion. The seawater has the very high concentration of chloride compared to groundwater. Thus, it can be used to identify intrusion, *i.e.* increased chloride concentration in groundwater (Mongelli *et al.*, 2013).

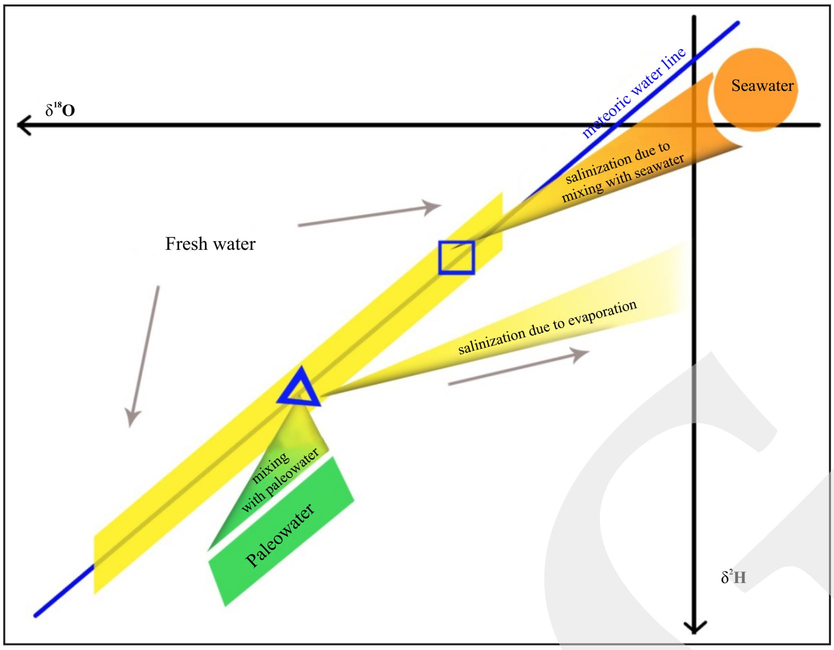

Figure 2. Graph of $\delta^{18}O$ *vs*. $\delta^{2}H$ shows modification of water isotopic composition due to several physical processes (modified after Yurtsever, 1997).

Results

Water Chemistry

The result of chemical analysis is presented in Table 1. The charge balances of the analysis results are less than 10%, which represents a good analytical practice. A number of 33 samples have TDS of less than 1000 ppm, while 14 samples have TDS of up to 3500 ppm, and 2 samples (Sawah Besar and Tanah Mas) have TDS of about 10,000 ppm.

Piper Diagram

Figure 3 shows a distribution of Semarang shallow groundwater during a dry season using the Piper diagram. Generally, the plot shows a shifted pattern from fresh water or shallow water composition, *i.e.* $CaHCO_3$ end member, towards sea water composition, *i.e.* NaCl end member. Samples from coastal area *i.e.* Pondok, Sawah Besar, Tanah Mas, Terboyo, and Puri Anjosmoro, are classified as NaCl and NaCaCl types which are typical of brackish to salty water. These types of water samples indicate a salinization process that may have occurred because of seawater intrusion or just salty water accumulation in the formation. The analysis of ^{18}O and ^{2}H isotopes can help clarify the evolution of these waters.

On the plain area further from coast line, *i.e.* Tanggulsari, Krajan, Gayamsari, Genuk, Genuksari, Bidakan, Pedurungan, Tlogomulyo, and Pelan, the water types of shallow groundwater are $NaClHCO_3$, NaCaCl, and $NaCaClHCO_3$ types. They are typical of a mix between fresh water and salty water. Thus, low to medium salinization processes also occurred in these areas.

Stable Isotopes of ^{18}O and ^{2}H

The result of isotope analysis presented in Table 2 and Figure 4 shows the graph of $\delta^{18}O$ *vs.* $\delta^{2}H$ of Semarang shallow groundwater. It can be concluded that some of the samples were shifted from its original position and more enriched compared to typical groundwater composition. Based on this plot, the samples can be classified into three groups:

Freshwater

There are twenty-four samples (51%) of shallow groundwater that plotted around the meteoric water line and no significant isotopic shift. These

Table 1. Results of Chemical Analysis (in ppm unit)

No	Location	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	TDS	Charge balance (%)
1	Tanggulsari	163.00	14.20	9.40	5.10	191.00	28.50	134.80	546.0	1.87
2	Karang Nangka	52.10	8.30	43.30	28.00	51.83	84.40	183.60	451.5	5.85
3	Wonosari	90.60	10.10	64.40	41.00	122.80	64.50	308.50	701.9	5.06
4	Rowosari	57.38	5.33	51.57	43.70	64.68	47.50	338.30	608.5	3.00
5	Krajan	122.96	23.12	119.20	44.60	315.50	99.80	208.44	933.6	4.86
6	Pondok	471.90	20.30	90.80	67.40	682.50	172.00	313.50	1818.4	6.36
7	Margosari	35.90	4.50	39.50	16.20	32.30	58.70	145.60	332.7	5.22
8	Puspagiwang	63.60	19.00	74.60	9.90	42.80	42.30	307.70	559.9	4.71
9	Purwoyoso	49.80	10.10	45.30	21.90	36.10	37.20	287.23	487.6	0.20
10	Pasadena	30.80	3.10	43.20	9.60	17.00	40.30	163.00	307.0	4.73
11	Tugu Lapangan	22.50	7.50	49.40	13.30	21.10	56.00	158.00	327.8	4.46
12	Karanganyar	285.20	15.20	53.20	18.60	309.00	162.00	238.20	1081.4	3.79
13	Ngliwonan	38.20	8.30	53.30	14.20	49.90	47.70	171.20	382.8	5.01
14	Tugurejo	149.30	26.90	49.30	18.50	183.80	90.50	151.50	669.8	8.60
15	Tambakharjo	270.70	40.20	92.60	35.00	351.80	88.50	417.70	1296.5	5.16
16	Krapyak	109.70	7.00	82.00	25.10	138.80	65.40	248.10	676.1	9.29
17	Kendeng	47.40	10.00	56.40	11.60	42.80	56.00	195.20	419.4	4.78
18	Kalialang	78.80	4.60	133.60	11.40	78.00	92.90	328.40	727.7	8.29
19	Manyaran	15.60	6.80	35.60	13.30	23.80	14.20	134.00	243.3	8.54
20	Puri Anjosmoro	613.80	20.00	246.70	72.30	1451.20	135.50	352.50	2892.0	-3.04
21	Dorowati	156.00	13.30	60.40	15.40	250.65	27.30	200.20	723.3	3.16
22	Lemah Gempal	21.56	6.05	47.56	20.70	21.23	43.60	200.17	360.9	4.12
23	Lempongsari	35.26	5.00	40.86	7.42	33.93	30.40	149.70	302.6	3.60
24	Tengger	18.04	4.17	21.92	7.20	14.93	41.90	82.40	190.6	-0.97
25	Deliksari	80.60	9.70	55.00	23.40	60.60	28.50	344.90	602.7	3.24
26	Bongsari	56.50	8.05	52.40	12.10	55.10	58.00	180.32	422.5	5.09
27	Bidakan	165.80	31.20	116.80	17.80	272.60	65.40	301.90	971.5	5.29
28	Tanah Mas	3424.00	217.00	179.70	332.20	5577.00	203.70	759.30	10692.9	5.94
29	Kepatihan	461.00	20.48	220.50	7.30	710.00	133.90	590.00	2143.2	0.44
30	Petelan	146.80	17.80	58.70	21.10	195.00	98.00	246.50	783.9	0.40
31	Wonodri	45.90	16.20	46.70	9.90	41.60	38.00	143.90	392.2	4.25
32	Candisari	33.90	6.48	30.88	14.20	32.10	35.60	170.40	323.6	-0.65
33	Jangli	43.60	9.80	45.30	9.20	46.80	57.40	147.20	359.3	2.79
34	Terboyo	927.40	34.20	172.60	87.50	1647.00	149.20	438.30	3456.2	1.46
35	Sawah Besar	3005.30	110.50	284.80	277.70	4991.90	409.50	384.60	9464.3	5.92
36	Sambirejo	231.70	16.20	64.60	45.00	325.00	70.70	359.90	1113.1	3.44
37	Gayamsari	126.90	15.60	44.70	15.20	166.50	68.80	200.10	637.8	0.71
38	Sengdangguo	66.80	9.50	33.97	11.10	52.24	55.80	171.20	400.6	3.26
39	Amposari	51.90	11.70	63.20	14.70	44.80	81.10	173.70	441.1	9.19
40	Juwono	15.76	13.30	35.51	14.20	24.85	24.80	165.40	293.8	0.84
41	Genuk	209.00	15.40	110.00	8.00	271.70	115.00	285.00	1014.1	3.75
42	Karangroto	325.50	14.60	95.40	8.70	450.90	115.60	324.25	1335.0	-0.15
43	Genuksari	288.90	7.30	76.30	15.20	390.90	64.90	250.00	1093.5	4.88
44	Tlogorejo	279.30	32.40	190.20	73.00	480.60	350.00	400.30	1805.8	2.69
45	Pedurungan	134.60	7.70	86.60	16.20	197.30	88.50	267.30	798.2	0.37
46	Penggaron	82.40	14.10	107.40	29.90	54.70	70.40	483.90	842.8	3.91
47	Tlogomulyo	394.10	23.40	66.60	88.20	489.30	294.00	367.30	1722.9	5.20
48	Godean	273.40	14.00	151.50	24.40	362.90	121.90	484.70	1432.8	3.33
49	Meteseh	18.40	5.98	63.80	8.40	21.00	27.40	271.31	416.3	-7.25

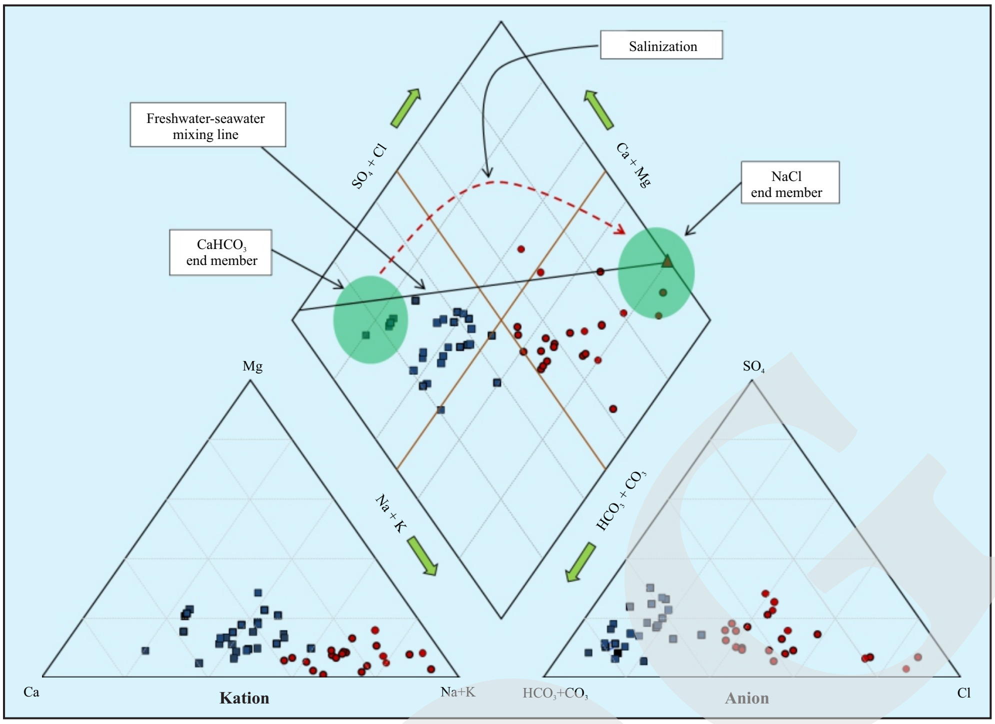

Figure 3. Piper diagram of Semarang shallow groundwater (blue squares = fresh water, red circles = mixing or ionic exchange).

Table 2. Isotopes Analysis Results of Semarang Shallow Groundwater (in ‰)

No.	Location	$\delta^{18}O$	$\delta^{2}H$	No.	Location	$\delta^{18}O$	$\delta^{2}H$
1	Tanggulsari	-3.97	-24.3	27	Bidakan	-4.20	-30.0
2	Karangnangka	-6.66	-36.9	28	Tanah Mas	-1.37	-16.0
3	Wonosari	-5.51	-31.7	29	Kepatihan	-3.29	-21.7
4	Rowosari	-5.47	-33.9	30	Petelan	-4.08	-26.5
5	Krajan	-4.04	-31.7	31	Wonodri	-6.30	-34.2
6	Pondok	-3.65	-30.8	32	Candisari	-6.31	-34.9
7	Margosari	-6.65	-35.3	33	Jangli	-6.35	-35.9
8	Puspa Giwang	-6.65	-37.2	34	Terboyo	-3.09	-23.6
9	Purwoyoso	-6.57	-35.4	35	SawahBesar	-1.32	-17.4
10	Pasadena	-5.92	-32.2	36	Sambi Rejo	-4.17	-29.5
11	TuguLapangan	-5.29	-34.5	37	Gayam Sari	-4.89	-30.7
12	Karang Anyar	-3.97	-29.3	38	Sendang Guo	-5.71	-35.3
13	Ngliwonan	-6.43	-33.0	39	Amposari	-5.19	-31.7
14	Tugurejo	-4.50	-32.0	40	Juwono	-5.87	-30.4
15	Tambak Harjo	-4.01	-30.5	41	Genuk	-3.87	-26.2
16	Krakyak	-4.93	31.0	42	Karangroto	-4.02	-32.4
17	Kendeng	-5.41	-32.5	43	Genuk Sari	-4.15	-27.7
18	Kalialang	-6.04	-34.3	44	Tlogorejo	-3.98	-28.0
19	Manyaran	-6.50	-35.6	45	Pedurungan	-4.83	-31.8
20	Puri Anjosmoro	-2.93	-21.8	46	Penggaron	-5.65	-30.0
21	Dorowati	-4.43	-27.7	47	Tlogomulyo	-4.44	-30.4
22	LemahGempal	-5.47	-34.3	48	Godean	-4.70	-32.9
23	Lempong Sari	-5.24	-34.3	49	Meteseh	-6.30	-30.8
24	Tengger	-5.75	-35.7	50	AL MarinaSemarang	0.45	-3.1
25	Deliksari	-5.39	-31.1	51	AL Pelabuhan	0.36	-1.2
26	Bongsari	-5.87	-32.9	52	AL Maron	0.36	-2.2

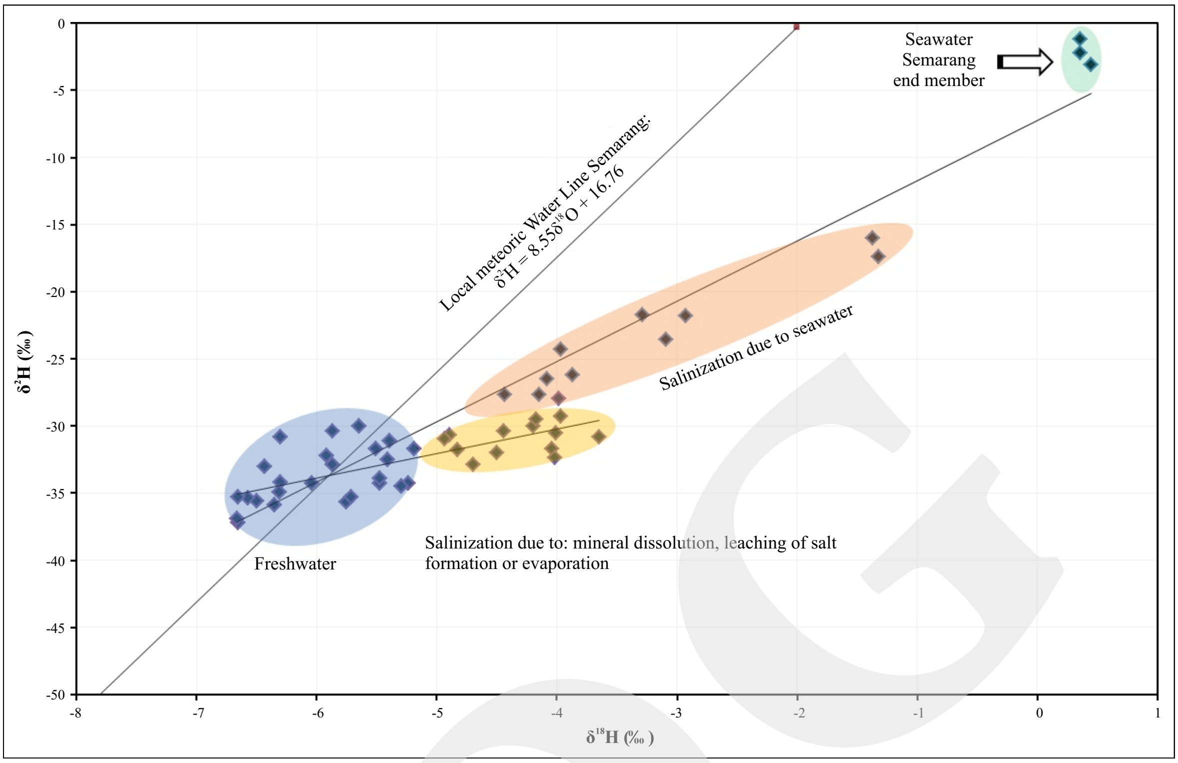

Figure 4. Graphic of $\delta^{18}O$ *vs.* $\delta^{2}H$ of Semarang shallow groundwater.

samples are taken from Rowosari, Wonosari, Ngliwonan, Manyaran, Jangli, Juwono, and Meteseh. These areas are located between 15 m to 142 m above sea level, which topographically are foot hills and hilly areas composed of volcanic rocks as sand gravels on top and breccias underneath.

Salinization due to mineral dissolution, leaching of salt formations or evaporation

There are 13 (27%) of shallow groundwater in this group which have isotopic shift from its original composition as if evaporation phenomenon. However, from its chemical composition as shown in the Piper diagram, these samples indicated the occurrence of a salinization process which is more likely than evaporation. The salinization process is probably caused by leaching of salt formations or mineral dissolution (Yurtsever, 1997). This phenomenon occurs in Pondok, Krajan, Karang Anyar, Tugurejo, Tambak Harjo, Bidakan, Sambirejo, Karangroto, Gayamsari, Pedurungan, Tlogomulyo, Godean, and Krapyak.

Salinization due to seawater intrusion

The rest of the samples, *i.e.* 11 (22%) are plotted between fresh water composition and seawater composition which indicates a mix with seawater with different proportion. Areas with low salinization are located in Tanggulsari, Genuksari, Genuk, Dorowati, Petelan, and Tlogorejo, with $\delta^{18}O$ composition between -4.43‰ and -3.87‰ and for $\delta^{2}H$ between -28.0‰ and -24.3‰. Areas with medium salinization are located in Puri Anjosmoro, Kepatihan, and Terboyo, with $\delta^{18}O$ composition between -3.29‰ and -2.93‰ and for $\delta^{2}H$ between -23.6‰ and -21.7‰. Areas with high salinization are located in Sawah Besar and Tanah Mas, with $\delta^{18}O$ composition between -1.37‰ and -1.32‰ and for $\delta^{2}H$ between -17.4 and -16.0‰.

Distribution of ^{18}O

The distribution of $\delta^{18}O$ value can be visualized using isocontour plot (Figure 5). Thus, isotopic shift pattern can be recognized geographically from a coastal area to a hilly area. Generally, sea-

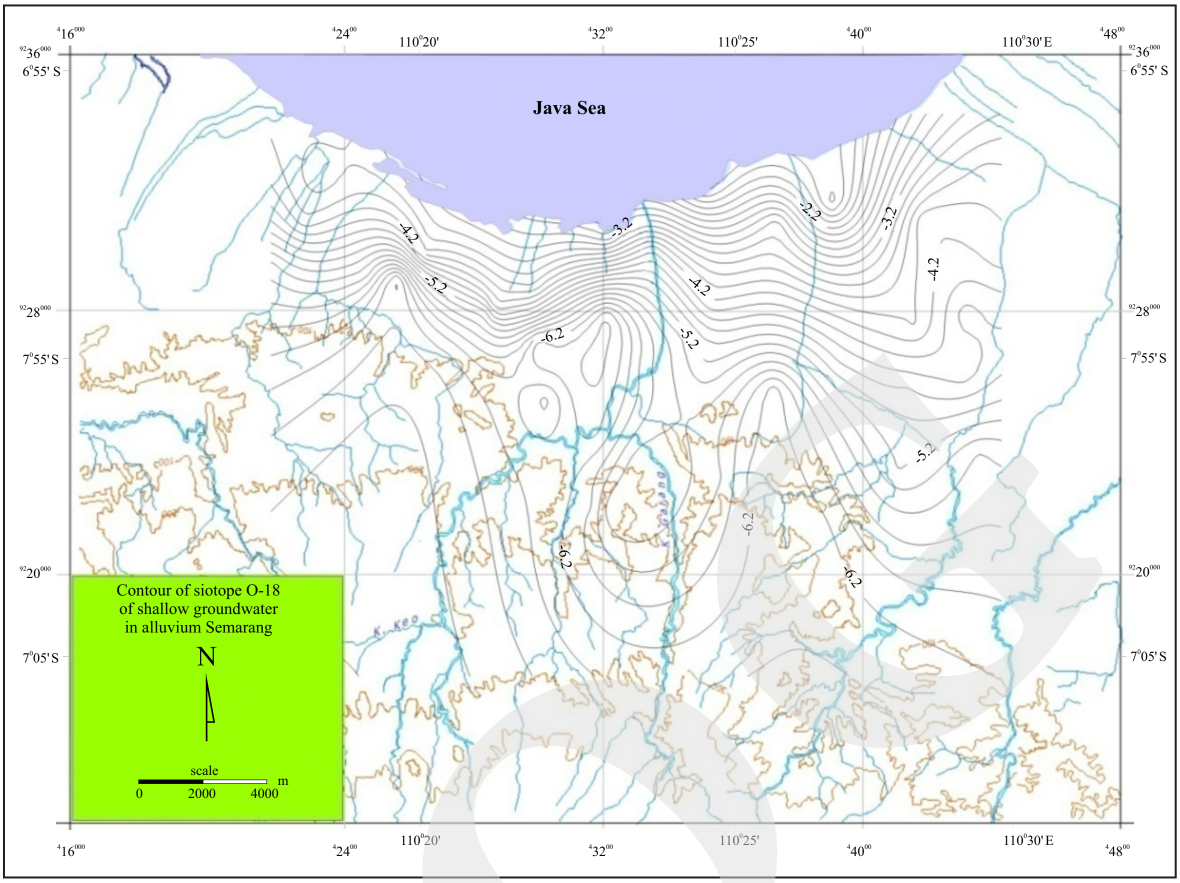

Figure 5. Contour of δ[18]O distribution in shallow groundwater of Semarang area.

water has δ[18]O composition of around 0 ‰. While Semarang seawater δ[18]O compositions are slightly more enriched, *i.e.* between 0.33‰ and 0.45‰. The seawater intrusion changes the isotope composition of groundwater into more enriched or more positive value. Based on the contour, the δ[18]O shift pattern from East Semarang about 1 km inland from coastline, the δ[18]O composition varied from 1‰ to 2‰ and for every 1 km to the south there is -1‰ shift. While in the West Semarang area, the δ[18]O composition 1 km from coast line are varied from -2‰ to -3‰ and for every 1 km to the south there is -1‰ shift. From this evidence, it is estimated that seawater intrusion has reach 4 - 6 km inland in the east of Semarang and 1 - 3 inland in the west of Semarang.

The δ[18]O distribution pattern also shows that in the centre of Semarang, the contour deflects to the south, *i.e.* hilly areas. This phenomenon is probably due to the existence of a fault zone or an existence of an impermeable zone that caused the groundwater flow deflected into a more permeable zone.

Conclusion

Based on the hydrochemistry analysis, shallow groundwater in Semarang mainly consists of $Ca-HCO_3$ type freshwater which spread from foot hills to hilly area in the southern of Semarang. While the other types are brackish to salty water of $Na-HCO_3-Cl$ type and Na-Cl type spreading along the coastline. Typical water with high Cl content generally occurs along the coastline as an impact of a salinization process.

The values of δ[18]O and δ[2]H show that shallow groundwater of Semarang is classified into three groups, *i.e.* freshwater, saline water due to mineral dissolution, and saline water due to seawater intrusion. In comparison with local meteoric line, all δ[18]O and δ[2]H values of freshwater in the studied area are plotted near the line which indicates its meteoric origin. Whereas saline groundwaters are more enriched in composition and plotted between freshwater and seawater composition, that indicates a mix between fresh and seawater.

Isocontour pattern of $\delta^{18}O$ shows the enriched values in groundwater distributed 1 to 5 km from coastline, *i.e.* -1 ‰ to -3 ‰, which are close to seawater composition. This phenomenon indicates that salinization process has reached 5 km inland. Thus, this pattern indicates that salinization due to seawater intrusion has reached 1 - 3 km inland in western area of Semarang, while for eastern Semarang reached 3 - 5 km inland.

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