Semarang, and its underground problems: Spatiotemporal hydrochemical assessment of shallow groundwater in Semarang, Indonesia

A. San Llorente Capdevila[1,2,3,4], T. Stigter[2], M. Kooy[2]

[1]Diponegoro University (Universitas Diponegoro), Jawa Tengah 50275, Indonesia

[2]IHE – Institute for Water Education, Delft, Netherlands.

[3]CERIS, Instituto Superior Técnico, Lisboa. Portugal.

[4]TU Dresden, Faculty of Environmental Sciences, Dresden, Germany.

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ABSTRACT

This research analyses the unconfined aquifer of Semarang lowlands, since it is suspected to be impacted by the current rapid development of the city. By assessing the current state of the unconfined aquifer, via a hydrogeochemical and multivariate statistics analysis of the collected groundwater samples from Semarang lowlands in 2017 and 2019, and a survey on water uses and practices of the same dug well samples. It is identified two kinds of pollution: diffuse and point-source pollution. The first one is found in the vicinities of industrial areas, and is characterised by seawater intrusion. The latter is found in the form of nitrate and nitrite in some of the 30 dug wells, including E. coli is found in all 30 dug well samples, ranging from 1 to 5 MPN/L. Seawater intrusion is reflected in the water-type composition of dug wells, and there are two water types characterising Semarang lowlands: Ca-Na-HCO_3 (with varying concentrations of Ca, Na) and HCO3, (with a low concentration of Cl). PCA results not only evidence but support the previous findings on the hydrochemical variables which play a major role, and therefore are influential, to the groundwater quality of the area. Point source pollution is also evidenced in statistical analysis, where in the PCA results of 2019 show a correlation between nitrite and ammonium, but this is different for 2017 results. The quality of groundwater has an influence on the uses and practices of the dug well owners of Semarang lowlands. By discarding the end user habit of using groundwater as a drinking source, and move to potable water sources such as bottled water, or treated water from the tap.

Keywords: Unconfined aquifer, water uses, seawater intrusion, nitrate pollution, Semarang, Indonesia

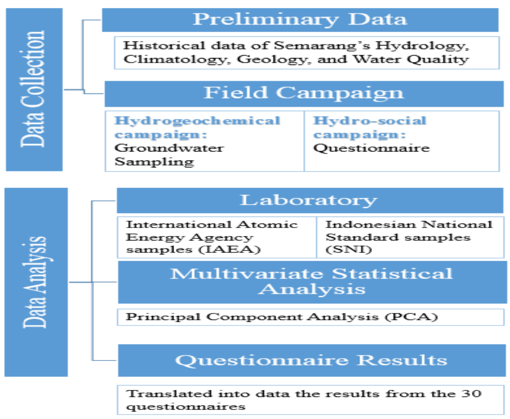
Background

The UN Environment publication on 'Progress of Ambient Water Quality' in 2018, observed that the achievement of Sustainable Development Goal (SDG) 6 related to ensuring availability and sustainable management of water and sanitation for all, is globally falling behind (SDG, 2011). This issue was further discussed in the High-Level Political Forum (HLPF) in 2018, and one of the important indicators to review SDG 6 was indicator 6.3.2: 'proportion of bodies of water with good ambient water quality'. To ensure clean water and sanitation for all by 2030, countries need to invest in adequate infrastructure, encourage hygiene, and good sanitation facilities (UNDP, 2019). The global population is at a constant growth from 3 billion in 1960 to 7.5 billion in 2018 (World Bank, 2018). Migratory movements follow this increase in population regionally and internationally towards coastal cities (Khumaedi and Purnomo Putro, 2017). Coastal areas exhibit a higher rate of population growth and urbanization but simultaneously correspond to areas of high vulnerability of the current climate change patterns (Neumann, *et al.*, 2015; Nganyi, Akrofi, Farmer, UNEP-GPA, WMO, and UNEP-WCMC, 2012). This population growth is increasing the pressure on resources and commodities of coastal ecosystems (Syvistski, 2008).

Coastal ecosystems naturally possess a saltwater – freshwater balanced interface where the land meets the sea. This balance can be easily disturbed by the local geomorphology, paleo-waters and weather events, tides, extreme-rainfall, or floods, and cause seepages of saltwater into the aquifer (Imam Wahyudi, *et al.*, 2017; Rahmawati, *et al.*, 2013). Furthermore, these areas are often found under pressing anthropogenic pressures, which also distort the natural fluctuations of the named interface, which cause an increment of saline intrusion into coastal aquifers. This leads to a decrease of groundwater quality for users of this source. Groundwater, once known to be the 'invisible' water source, has been moved over the last decade to make headlines by the economic and population growth worldwide (Kemper, 2004). This source is subject to vulnerability, which is enhanced by the increased stress on fresh water by the populations' growth and withdrawal of this source (Datta and Singh, 2014; Khumaedi and Purnomo Putro, 2017). This is putting at risk one of the most important renewable and natural resources, groundwater (Datta and Singh, 2014). Leading to the salinization of surface waters and shallow fresh groundwater bodies, making the water unfit for irrigation, drinking water supply or industrial purposes (DeLouw *et al.*, 2011; Worland, *et al.*, 2015). Where the supply of freshwater is not adequate, the forced consumption of saline water can impact on human health by promoting the development of renal failure, kidney disease, hypertension and gastrointestinal irritation (He and MacGregor, 2009).

Groundwater quality is often altered by more than a single pollutant. This is of major concern in water management for most of the world's developing areas (Boy-Roura, 2013; Sadler *et al.*, 2016). Aside from the degradation of water quality of aquifers by seawater intrusion, and other chemical pollutants such as nitrates responsible for the contamination of groundwater. Nitrates and nitrites are often to be by-products from agricultural and wastewater practices (Andrade and Stigter, 2009; Foster, 1987; Stigter, 2008; Templeton *et al.*, 2015). The degradation of quality of aquifers is largely blamed to the ongoing farming activities since the mid20th century, and other sources which introduce large quantities of nutrients, such as nitrates (Burg and Heaton, 1998; Buzek *et al.*, 1998; Dietrich and Hebert, 1997; Focazio *et al.*, 1998; Foster, 1987; OECD, 2008; Stigter *et al.*, 2006). The other contributors are the discharge from leaking sewers, septic tanks, spreading of sewage sludge and atmospheric deposition (Mtoni *et al.*, 2013; Templeton *et al.*, 2015; Stigter *et al*, 2008; Wakida and Lerner, 2005).The artificially added nutrient-load by these activities is often found to be harmful for human health, the *WHO* since 2008 in their studies that the ingestion of nitrate even below their guidelines (50mg/L) has a high risk of contracting certain cancers, adverse pregnancy outcomes, diabetes and thyroid disorders (Boy-Roura, 2013; Ward and Brender, 2011; WHO, 51 2008; Ward *et al.*, 2005). Other studies have expressed their concern for human health exposed to the small quantities of nitrates and nitrites, since research has mainly focussed on the extensive study for nitrite concentrations above guidelines levels (Philips, *et al.*, 2002). Furthermore, the scientific community has been growing more concern towards the environmental problems which stem from the increasing number of nitrates as these nutrients are very difficult to remove from surface fresh- and salt- waters that cause the acceleration of algae growth leading to the eutrophication of environments (EC, 1991; Stigter *et al.*, 2006).

Materials and Methodologies



Scheme 1. Graphical representation of the methodology applied in this master thesis.

1.1 Methodologies for field data collection

1.1.1 Hydrogeochemical campaign

This sampling campaign is comprised by the physical and the chemical sampling of the groundwater from dug-wells in Semarang City. In order to understand the water quality of this study, is of high importance to identify its parameters. These defining practices are water quality are physical, chemical, and biological indicators using indicators such as pH, electrical conductivity (EC), total dissolved solids (TDS), hardness, turbidity and contaminant concentrations based on guidelines provided by agencies such as the World Health Organization WHO (2006) and the Bureau of Indonesian Standards (BSN) (Mahapatra et al., 2012).

The physical and chemical parameters sampled during this campaign are detailed in *table 1.2.1-1.* The followed sampling protocol for E. coli was given by the Public Health Department of University of Diponegoro (UNDIP). From now on cited in this research as UNDIP. This protocol fits the sampling capacities of the university, following a similar methodology for the groundwater sampling by Bordner *et al.,* (1978), Environmental Protection Agency (EPA) (2002), and Harter *et al.,* 2014. The hydrogeochemical sampling campaign followed two different protocols. The first one is the groundwater sampling protocol by the International Atomic of Energy Agency (IAEA), and the second one is the groundwater sampling protocol by the national standard of Indonesia (SNI). The latter method was applied in the 2019 campaign and in the first field campaign in 2017 by the University of Diponegoro (UNDIP). IAEA protocol was only applied in 2019. SNI protocol differs from IAEA by its refrain to collecting 500mL water samples from each dug well (without filtering water), using an opaque plastic water bottle (not acidified), and kept at room temperature. Since the 2017 campaign was sampled using the SNI protocol, this study wanted to test for differences in their respective results. In the sampling campaign of 2019, the two types of dug well samples were processed differently in the lab following its respective protocol, details found in *Section 4.1*.

1.1.2 Hydro-social data collection

The second field campaign was carried out for a deeper understanding of water uses. Via the previously mentioned voluntary questionnaire to all household owners of dug-wells. The aim of this questionnaire is to obtain an understanding of the current sanitary and clean water situation of Semarang City. At the same time as understanding the local knowledge on groundwater, water uses and practices.

1.2 Laboratory analyses

1.2.1 Hydrogeochemical laboratory analysis

The groundwater samples were analysed in the laboratory of "Laboratorium Mekanika Tanah and Batuan PAG Badan Geologi". As previously explained the IAEA and SNI sample groups where analysed differently in the laboratory. SNI had a turbidity test, before moving onto the filtration and preservation via acidification to perform the analysis of ions. This protocol establishes a max period of storage of 2 weeks. IAEA samples were analysed directly with no pre-treatment. Both samples analysed following the same laboratory methods, *table 1.2.1-1.*

Measurement	Laboratory procedure
K^+, Na^+, Li^+, Ca^{2+}, Mg^{2+}	Ion Chromatography
Fe2+, Mn^{2+}	Atom Absorption Spectrophotometry
HCO_3^-	Volumteri (Alkalinity Test/Titrimetric)
Cl^-	Volumetri (Argentometri/Titrimetric)
SO_4^{2-}, NO_2^-, NO_3^-, NH_4^+	Spectrophotometry
pH	Potensiometry
EC	Conductometry
Colour	Spectrometry
Turbidity	Turbiditymetry
E. Coli	Dilution method

Table 1.2.1-1 List of the different laboratory procedures applied for the analysis of the groundwater samples of Semarang Lowlands.

1.2.2 Bacterial (E. coli) laboratory analysis

Following EPA's guidelines (2002) and SNI 01-2332.1-2006, *E. coli* samples were analysed as after its collection. This research did not hold these samples for longer than 6 hours, because of holding time limitations. Analyses on the samples were finished within 8 hours of sample collection. *E. coli* samples were analysed by the Most Probable Number (MPN) method following the dilution method by *Salamat et al.,* (1978). The coliforms were tested for by inoculation of samples into tubes of lactose broth by Collins and Lyne (1976). Find the results and WHO table in *Annex II.*

1.3 Data analyses

1.3.1 Hydrogeochemical data

The results from the laboratory analysis were plotted with ArcGIS and QGIS, to visualize the variation in time of the chemical components of the groundwater of Semarang lowlands. By making use of the geographic information, and the interpolation of data points to produce contour maps, and the distribution of the physical and chemical parameters.

Prior to the analysing of the laboratory results, this study performed the Electro Negativity (EN) principle to test for the reliability of the samples. This method calculates the ion balance errors of each sample, *Equation 4.5.1-1*. This principal says that water is not able to carry a net electrical charge, being positive or negative, but must always be electrically neutral. Since most dissolved species carry a charge, electronegativity demands that the sum of equivalents of positively charged species matches the sum of equivalents of negatively charged species (Appelo and Postma, 2005).

$$E.N(\%) = \frac{\sum cations + \sum anions}{\sum cations - \sum anions}\, x\ 100$$

Equation 1.3.1-1 Electronegativity principle equation.

1.3.2 Multivariate analyses of Hydrogeochemical and physical parameters

This study used a multivariate statistical analysis for the groundwater samples, to identify the controlling processes of the unconfined aquifer of Semarang City. These statistical analyses used the data from the field campaign. This chemical data is then presented in graphical form to obtain a simpler understanding of the complex groundwater system. The methods used to represent the groundwater chemistry are Piper plot, Gibbs plot, Schöeller, and Principal Component Analysis (PCA), which from now on Principal Component Analyses will be referred as PCA in this thesis. These methods have been used in many parts of the world to show the latent relationships amongst the various ionic concentrations in individual samples, and are making use of these statistics in the work with the same objective (Pan and Ritcher, 2019). The analysed variables using this PCA are detailed in *table1.7-1.*

1.3.3 Escherichia Coli (E. Coli) and Nitrogen components analysis

The results from the laboratory analyses of E. coli and Nitrogen compounds nitrate/nitrite were analysed by visualising the spatial distribution of 2017 and 2019 results through the software ArcGIS (Osrmby *et al.*, 2010). Global water guidelines of WHO were used for reference and scaling (WHO, 2015).

1.3.4 Hydro-social campaign qualitative analysis

This qualitative analysis was based on the 30 interviews to dug-wells owners that will be carried out during the fieldwork in Semarang. Results

Results

1.4 Natural groundwater chemical processes and evolution

Similarities and differences among groundwater samples are revealed through the Piper diagram, *figure 1.4-1*. Within these triangles the distribution of major cations and anions of the 30 dug well samples is observed for 2017 and 2019. These samples are then combined and illustrated within the diamond shape area (Appelo and Postma, 2005). These combinations will be representing the main groundwater facies found in Semarang lowlands. Piper diagram results shows a high dominance in cations of Ca, and Na, and in anions of HCO_3 and Cl for both years. In both years, the data clusters over the groundwater type Ca-HCO_3 and Na-HCO_3 for fresher waters (<1500 µS/cm), and spreads to right side of the diamond shape, reaching towards the groundwater facies of Na-Cl and Na-HCO_3 for more brackish waters (>1500 µS/cm). These 60 dug well samples fall within the groundwater facies of: Ca-HCO_3, Ca-Cl, Na-HCO_3 and Na-Cl. From these findings it is understood that in some dug wells there is some seawater intrusion in Semarang lowlands.

The differences between the more groundwater facies of brackish samples arise when comparing the dug wells samples from 2019 (light blue) with the 2017 (navy blue) ones. In 2019, brackish samples cluster on the lower right part of the diamond shape, showing up in the groundwater facies of Na-Cl and Na-HCO_3. In 2017, brackish samples are more disperse across the right side of the diamond shape. They appear to move from Ca-HCO_3 to Na-Cl and Na-HCO_3 water types. This is indicative to the brackish dug well samples of 2019 and 2017 to be under a different influence. Since 2019 appears to be more abundant in sodium compared to 2017, which shows to have less sodium and more calcium in its water types.

Freshwater samples of 2019 and 2017 differ between years. In white it shows the 2019 samples, which are mostly present on the lower right part of the diamond belonging to the water type Na-HCO_3 and Na-Cl. Differently in 2017 freshwater samples are more clustered on the left-hand side of the diamond shape, within the water type of Ca-HCO_3. Again, the results show a different mineralizatrion for groundwaters, which may be understood as having different influences in each year and therefore other processes may be ocurring, i.e. freshening. In order to reach clearer conclusions, this study takes these differences to a deeper analysis with bivariate plots, *figure 1.5-1.*

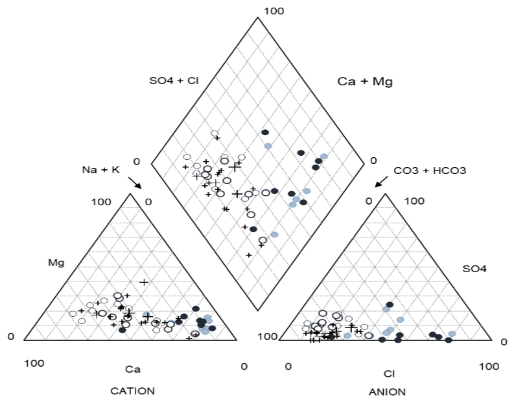


Figure 1.4-1 Piper graph of 2019 and 2017 sampling campaigns in Semarang Lowlands. The blue colours represent samples >1500 µS/cm, being the light blue 2019 samples and the navy blue 2017. The white and black symbols represent samples <1500 µS/cm, in white for 2019 and in black (+) for 2017.

1.5 Major and minor ions results

Figure 1.5-1 shows the sodium vs chloride concentrations for 2019 and 2017. In this figure the samples of this study are compared to pure seawater in order to check whether they fit along the mixing line. Most samples seem to plot above the conservative mixing line, indicating a higher concentration of Na over Cl. With an exception of 6 samples which are the previously observed 6 dug wells that deviate from the norm in Gibbs and Piper graphs, and are neighbouring industrial areas. These samples may be polluted by seawater intrusion, which will be further discussed in *Chapter 6*. In the following figures for 2019 and 2017, *figure 1.5-1b,* shows all samples except one to be plotting above the 0.86 ratio. This figure only displays the ratio of the samples of this study. As getting closer to the marine ratio the concentration of sodium seems to decrease as chloride concentration increase. The relationship between Na and Cl is also highlighted in the Pearson results, as they appear to be highly correlated in both sampling years, in 2019 r = 0.96 and in 2017 r = 0.98.

Figure 1.5-1d, shows both years with an increase in alkalinity through the dug wells, and some sort of linear relationship, which does not appear in the Pearson's results of 2019 and 2017, r = 0.19 and 0.28, respectively. SG25 is the one found in both years to have the most extreme values, with the highest concentration of bicarbonate: 14.07 meq/L in 2019, and 17.07 meq/L in 2017. The dug wells found on the negative side of the y-axis in both years when adding calcium, they become part of the highest samples. Indicating that they have a great contribution of calcium in their composition. These samples were found close to industrial areas for both years. In 2017 shows that the samples above 12 meq/L are SG1, 20, 21, 24, 25, 27, and 28, and are under suspicion of seawater intrusion. In 2019 shows a smaller concentration of bicarbonate compared to 2017, and a single sample SG25 above the cut-off point of 12 meq/L. When this research uses 10 meq/L as cut of point, it results with 5 samples exceeding it: SG4, 16, 24, 25, and 27. Differently to 2017, all these dug wells samples belong to the 8 dug well group with potential of seawater intrusion. *Figure 1.5-1c* displays an increase of salinity and alkalinity through the samples. These 6 previously spotted are the dug wells plotting furthest to the right. These have a higher concentration of chloride compared to the most groundwater samples. The concentrations of bicarbonate (HCO_3) and calcium (Ca), in *Figure 1.5-1d*, show the dug wells are found under the dissolution line of $CaCO_3$, which correspond to the Pearson correlation results of r = 0.19 in 2019 and r = 0.28 in 2017. This positive correlation shows salinity is not recent, where Cl would substitute HCO_3 it seems that Cl can originate from entrapped seawater or from a common source; wastewater or evapotranspiration, further discussed in *section Discussion 6.2.2*. *Figure 1.5-1g* illustrates little variation in concentrations of Sulphate (SO_4) and Chloride (Cl) between the sampling campaigns of 2019 and 2017. In both years there seems to have a positive correlation between the two anions. This is evidenced though the Pearson analysis showing r = 0.59 in 2019 and r = 0.63 in 2017. All samples appear to be above the ocean ratio (SO4/Cl), in 2019 most samples lie between the 0 - 1% with a few samples reaching a 5%. Differently to 2019, in the results of 2017 appear to be at lower values, and are mostly found to be under the mixing line. Most samples lie between 0-1% with a few reaching the 4% maximum, indicating sulphate dominates over chloride in most samples. This is interpreted as a possible indication of more oxidizing conditions in 2019 compared to 2017. The relationship between sulphate and nitrate is further looked into in *figure 1.5-2.*

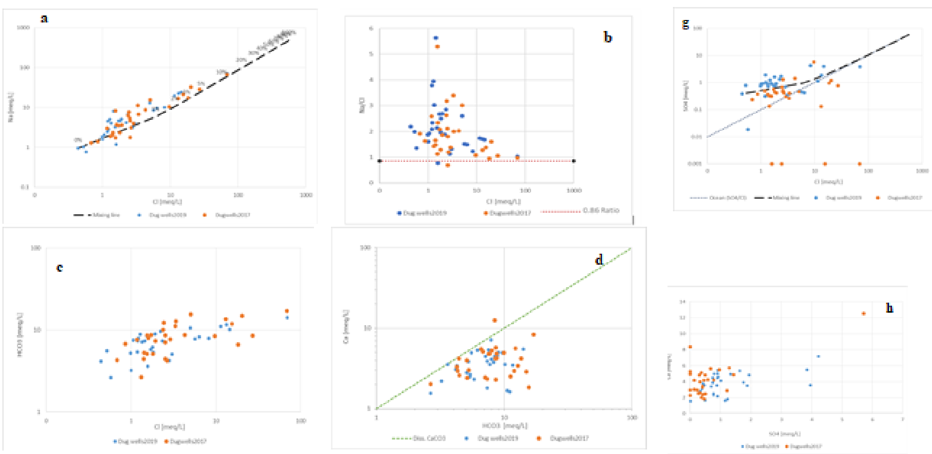


Figure 1.5-1 Scatter plots for the major ions per unit in meq/L for the 30 dug well samples from Semarang lowlands 2019 (blue) and 2017 (orange). Navy dotted lines indicate the ocean ratio of concentrations. Mixing lines are represented in the respective colours for each year in a line with a marker. This illustrates the conservative mixing between Appelo and Postma (2005) seawater endmember and a freshwater endmember (SG14). Lines with round markers represent the percentage contribution of seawater in steps of 1, 2, 3, 5, 10, 20, 30, 40, 50, 60, 80 and 90.

Figure 1.5-2a shows the relationship between the saturation indexes of sulphate against nitrate for 2019 and 2017. These results were obtained through the software of Phreeqc, and they illustrate a very different behaviour between the two components in in both years. In 2019 a great number of samples are clustered on the left side of the graph, between 0 – 0.1 NO_3 mmol/L and in 2017 samples are clustered at the bottom of the graph, never exceeding the 0.7 NO_3 mmol/L in exception of one sample: SG25. In this figure there are a number of samples identified as suspicious (inside a red circle) and further analysed in the *Chapter 6 Discussion. Figure 1.5-2b* displays the relationship between pH and pCO_2 in 2019 and 2017. Similarly, this figure shows a difference in behaviour of groundwater between the sampling years. In 2017 dug wells fall between the range -1 and -1.5 pCO_2, whereas in 2019 dug wells reach a wider range from -1.25 down to -2.75. This can be interpreted as reduction condition and again is a suspicious behaviour (highlighted in red) which will be taken to further discussion in *Chapter 6 Discussion. Figure 1.5-2c* displays the Saturation Index of calcite versus calcium. It displays the concentrations of these components for both sampling years. This time concentrations diverse mildly when compared to *figure 1.5-2a&b*, however 2017 has a wider range when compared to 2019. At the same time, in red, it is identified some suspicious behaviour which will be further discussed in *Chapter 6 Discussion.*

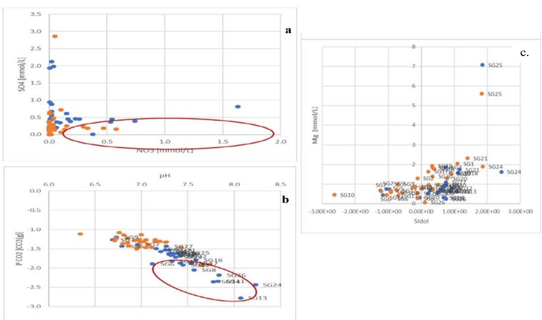


Figure 1.5-2 Scatter plots of Saturation Indexes of calcite (SIcc), nitrate (NO3), and Carbon dioxide pressure (pCO2) for 2017(orange) and 2019(blue) sampling campaigns of groundwater in Semarang lowlands. Dug wells are labelled within the figure (SGX). Circles indicate detected anomalies.

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1.6 Pearson correlation coefficient (r)

Table 1.6-1 shows the results for Pearson analysis on the 2019 dug well samples. This matrix shows how various chemical parameters from the sampled 30 dug wells are correlated. Some of these relationships have already been identified in the bivariate plots in *section 1.5*. *Table 1.6-1* shows the results for Pearson analysis on the 2017 dug well samples. In this correlation matrix there are different pairs to 2019 results, which are being significantly correlated, SO_4^{2-} and Ca^{2+}, and two similar pairs, SO_4^{2-} and Cl^-, NO_2^- and Fe^{3+}, and NO_3^- and EC. The first pair might be correlated due to sulphate reduction. The second pair might be indicative of cation exchange, and the third pair might be subject to the change of groundwater chemistry caused by anthropogenic influences. In this year, magnesium is significantly correlated to Mn^{2+}, K^+, Na^+, HCO_3^-, Cl^- pH, and TDS. Manganese is significantly positive correlated to K^+, Na^+, HCO_3^-, Cl^- pH, and TDS. Potassium is significantly correlated to Na^+, HCO_3^-, Cl^-, SO_4^{2-}, pH and TDS. Further studied in the *Discussion.*

	Ca^{2+}	Mg^{2+}	Fe^{2+}	Mn^{2+}	K^+	Na^+	NH_4^+	AlkL	Cl^-	SO_4^{2-}	NO_3^-	NO_2^-	Ecol	EC	pH	TDS
Ca^{2+}	1															
Mg^{2+}	0.47	1														
Fe^{2+}	0.23	0.21	1													
Mn^{2+}	0.35	0.50	0.20	1												
K^+	0.05	0.09	0.15	0.11	1											
Na^+	-0.15	0.29	0.00	0.20	0.63	1										
NH_4^+	-0.11	-0.11	0.21	0.14	0.13	0.07	1									
AlkL	0.19	0.52	0.28	0.20	0.56	0.76	0.12	1								
Cl^-	-0.05	0.38	0.01	0.32	0.59	0.86	0.09	0.65	1							
SO_4^{2-}	0.48	0.47	-0.08	0.43	0.40	0.55	-0.09	0.41	0.59	1						
NO_3^-	-0.03	0.13	0.24	-0.08	-0.04	0.05	0.54	0.09	0.07	-0.10	1					
NO_2^-	0.22	-0.09	-0.15	-0.28	0.04	-0.25	-0.27	-0.32	-0.21	0.00	-0.02	1				
Ecol	0.01	0.29	-0.03	0.10	0.12	0.30	0.26	0.09	0.38	0.21	0.26	0.15	1			
EC	-0.07	0.35	0.11	0.30	0.49	0.73	0.20	0.74	0.68	0.23	0.04	-0.21	0.33	1		
pH	-0.38	0.16	0.27	-0.01	0.26	0.54	0.16	0.53	0.41	0.08	0.34	-0.27	0.07	0.47	1	
TDS	-0.07	0.35	0.11	0.30	0.49	0.73	0.20	0.74	0.68	0.23	0.04	-0.21	0.33	1.00	0.47	1

	Ca^{2+}	Mg^{2+}	Fe^{2+}	Mn^{2+}	K^+	Na^+	NH_4^+	AlkL	Cl^-	SO_4^{2-}	NO_3^-	NO_2^-	EC	pH	TDS
Ca^{2+}	1														
Mg^{2+}	0.02	1													
Fe^{2+}	-0.06	0.19	1												
Mn^{2+}	0.02	1.00	0.20	1											
K^+	0.16	0.50	-0.09	0.51	1										
Na^+	0.01	0.56	-0.06	0.56	0.68	1									
NH_4^+	-0.08	0.04	0.30	0.04	0.13	-0.04	1								
AlkL	0.05	0.56	-0.11	0.56	0.58	0.65	0.21	1							
Cl^-	0.14	0.57	-0.01	0.57	0.69	0.93	-0.05	0.40	1						
SO_4^{2-}	0.82	0.05	0.17	0.05	0.07	0.16	-0.03	0.05	0.23	1					
NO_3^-	0.24	0.02	0.51	0.01	-0.09	-0.06	0.07	-0.19	0.05	0.35	1				
NO_2^-	-0.04	-0.20	-0.19	-0.20	-0.16	-0.30	-0.14	-0.40	-0.21	-0.07	-0.08	1			
EC	-0.23	0.39	0.01	0.39	0.29	0.56	0.06	0.69	0.35	-0.20	-0.18	-0.52	1		
pH	0.03	0.56	-0.11	0.56	0.58	0.65	0.21	1.00	0.40	0.05	-0.19	-0.40	0.69	1	
TDS	0.22	0.63	-0.05	0.63	0.74	0.97	0.02	0.64	0.95	0.30	0.01	-0.29	0.47	0.64	1

Table 1.6-1 Pearson correlation (r) matrix of 18 physico-chemical variables in groundwater samples from the Semarang lowlands above 2019 and below 2017. Numbers in red indicate significant correlation (r ≥0.5). AlkL stands for Alkalinity (HCO_3^-), Ecol for E. coli, EC, for Electrical Conductivity, and TDS for Total Dissolved Solids.

1.7 Principal Component Analysis (PCA)

Table 1.7-1 shows the PC groups for 2019. Highlighted in red *table 1.7-1* shows the parameters considered significant >Absolute (0.5). In PC1 there are nine parameters and they are grouped in three categories: (1) Physical: Electrical conductivity, pH, (2) Cations: Na+, K+, Mg2+, (3) Anions: HCO3-, Cl-, and SO42-. These parameters seem to be related and expressing the influence of the local geology in the groundwater processes. Another interesting relationship found in PC1 is the weak association of E. coli with all nitrogen compounds: NH4+, NO3-, NO2.

Variable	PC1	PC2	PC3	PC4	PC5
Ca^{2+}	0.12	-0.51	0.61	0.41	0.22
Mg^{2+}	0.75	-0.07	-0.06	-0.35	-0.23
Fe^{3+}	0.00	-0.15	0.36	-0.79	-0.10
Mn^{2+}	0.75	-0.08	-0.06	-0.35	-0.23
K^{+}	0.74	-0.23	-0.22	0.10	0.13
Na^{+}	0.91	-0.17	-0.16	0.09	-0.12
NH_4^{+}	0.08	0.07	-0.08	-0.33	0.78
AlkL	0.83	0.27	-0.10	0.03	0.34
Cl^{-}	0.81	-0.39	-0.10	0.07	-0.23
SO_4^{2-}	0.18	-0.58	0.64	0.22	0.21
NO_2^{-}	-0.07	-0.45	0.47	-0.52	0.05
NO_3^{-}	-0.47	-0.46	-0.48	0.15	-0.10
EC	0.64	0.42	-0.18	-0.15	0.09
pH	0.83	0.27	-0.10	0.03	0.34
TDS	0.93	-0.29	-0.05	0.10	-0.05

Table 1.7-1 The component matrix and principal components (PC) of 2019 sampling campaigns in Semarang Lowlands. In red are the considered significant coefficient │>0.5│. AlkL stands for Alkalinity (HCO_3^-), Ecol for E. coli, EC, for Electrical Conductivity, and TDS for Total Dissolved Solids

Discussion

Previous research explains that the groundwater in Semarang lowlands flows from South to the North of the city, where it meets the coastline (Lloyd et al., 1985; Putranto and Rüde, 2015). Groundwater is subject to atmospheric influences such as groundwater abstractions, wastewaters, evaporation or the local dry-deposited dust particles and gases, in other words, the local soil matrix (Appelo and Postma, 2005). In Semarang lowlands, Damar and Alluvium rock formations dominate the material that is being weathered (Abidin et al., 2010; Putranto and Rüde, 2005). This weathering of two key rock formations, along with oceanic influence, plays a major role in the groundwater chemistry of this study area, varying spatially; closer to highlands dugwells are influences by Damar formation and dug wells on the plains are influenced by the Alluvium formation. For example, figure 1.4-1 evidences that this study is dealing with groundwater at a discharge point, as there is a higher Na surplus in 2019, which seems consistent with earlier observations in the graph of *figure 1.5-1* (Appelo and Postma, 2005). Indicative to this study as it is also showing a source of sodium in Semarang lowlands, as the local geology, alluvium formation. Results showed Na at higher concentrations than Ca, indicating a more alkaline sample (figure 1.5-1d). This alkalinity may have a geological origin, since groundwater under volcanic influence (Damar) is characterized by silicate-type water, Ca-Na-HCO3, concurring with the Piper results. Ca and HCO3 ions result from calcite dissolution and silicate weathering (Appelo and Postma, 2005). Calcium carbonate is present in the alluvium formation in the form of forams, molluscs and coral colonies. *Putranto and Rüde* (2011) described "thick a layer of calcareous and shell bearing clay". There is no great difference in Ca concentrations between the two years, figure 1.5-1d, which means that the shift in the piper diagram is due to increased Na concentrations in 2019, *figure 1.4-1*. Another interpretation can be linked to the recharge rates, being in 2019 less than in 2017, and therefore older and more saturated waters are captured; finding Na+, HCO3 and Ca+2. The increase of sodium may originate from silicate weathering, which may be caused by an extreme weather event or an unusual discharge. The alluvium formation is composed by this calcium-rich layer with interlayers of clay layers composing the unconsolidated alluvium, along with siltstone or sandstone (Lloyd et al., 1985; Putranto and Rüde, 2011). Silicate weathering seems to play a more significant role in the groundwater of Semarang lowlands, although it is not repeated throughout all dug wells or in both sampling years. Both years present, more in 2017, dug wells which are undersaturated in Semarang lowlands, figure 1.5-1. This reveals younger waters, with high nitrate concentrations. Sulphate seems more reduced in 2017, and in overall, 2017 samples shift to higher salinities. The suspect of this are wastewaters from industrial areas. Nitrate is reduced, specially, and then nitrification seem to be occurring due to oxic conditions. pH is much lower and pCO2 is high, figure 1.5-2a,b,&c. This is indicative of an open dissolution system and reduction is causing high alkalinities, which high alkalinity is found at very high concentrations in 2017. These processes are also common paths of Cl- and HCO3, which can be caused by evapotranspiration or wastewater influence. Differently, in 2019 pCO2 is much lower and higher pH, this evidences a closed system dissolution compared to 2017. Further evidence towards the influence of local lithology to groundwater quality is found in the Mg vs Alkalinity (HCO3) relationship, table 1.6&7-1. A positive relationship between these variables can be observed. Strangely, the highest Mg concentration was found in dug well SG25, showing similar levels of Mg as in the coast of Semarang lowlands and in the middle of the industrial area.

Research by Putranto and Rüde (2011) could help explain the fluctuations in water-types from year to year, where they identified 2 groups in the alluvium aquifer; Garang aquifer and Quaternary marine aquifer. Garang aquifer is characterised by fresher waters compared to the more saline Quaternary marine aquifer (Haryadi et al., 1991; Sihwanto et al., 1988; Susana and Harnadi, 2007). The results of this study bring to light the fluctuations in the unconfined aquifer. The influence of local lithology to the groundwater quality is not only evidenced in the hydrogeochemical analysis of this research, but also in the statistical analysis of this thesis, section 5.3. Where PCA illustrates in its first principal component (PC1) the correlation between the components which characterise the local geology of Semarang lowlands; Mg+2, K+, Ca+2, Na+, and HCO3-. The correlation with Cl- shows the influence of more rich waters, probably older, as well as seawater intrusion in the sampled dugwells. These statistical evidences make all previous interpretations plausible.

Semarang City is under the risk of seawater intrusion, caused by the overexploitation of its aquifers (Irawan et al., 2018; Purnama and Marfai, 2012; Putranto and Rüde 2016; Rahmawati and Marfai 2013). This may contribute to the hypothesis that seawater intrusion influences the groundwater quality, a theory supported by the high concentrations of magnesium found in figure 1.5-2. This relationship between magnesium and alkalinity could be due to seawater mixing and possibly cation exchange. This is also identified in Pearson's analysis of 2017, where magnesium and chloride positively correlate. This statistical result supports this interpretation, as 2017 is the year with highest salinity. Previous research explains that Semarang lowlands geological evolution entrapped brackish water in the clay layers of the alluvium formation, where the unconfined aquifer is formed during the Pleistocene–Holocene ingressions caused by the climate changes occurring during the Quaternary (Purnama and Marfai, 2012; Putranto and Rüde 2016; Rahmawati and Marfai 2013). The content of major ions that predominate in sea water (Cl−, Na+ and Mg2+), demonstrates that such salinization is related to sea water (Morell and Gimenez, 1996; Gimenez and Morell, 1991). Also, the enrichment of Ca as the principal ion can be used as an indicator of seawater intrusion into groundwater (Gimenez and Morell, 1991; Morell and Gimenez, 1996; Somay and Gemici, 2009). The enrichment observed in the major ions (HCO3 and SO42-) with respect to the seawater mixture would show that the semi-confined aquifer water from the coastal plain corresponds to a paleowater intruding in the unconfined aquifer released from clay layers. This is also reflected in the positive correlation between the major ions, which can be interpreted as indicative of trapped seawater in clays, table 1.7-1. Figure 1.5-1g shows some of the dugwell samples have a deficit in sulphate. The concentrations of anions Cl and SO4 are very different between sampling years, figure 1.5-1g. In both years a positive correlation between the two anions can be observed, evidenced though the Pearson analysis, showing $r = 0.59$ in 2019 and $r = 0.63$ in 2017. All samples appear to be above the ocean ratio (SO4/Cl), and only a few go above 1 (figure 1.5-1g), further evidencing the sulphate deficit. The data shows sulphate concentrations to be much higher in 2019, with most samples falling between 0 - 1% and a few samples reaching a 5%. Sulphate, like NO3-N, is involved in biological processes and can be temporarily retained in soils or biological materials. The many sulphate trends found during this study illustrates the ion's mobility, and iron acting as an intermediate process (Appelo and Postma, 2005). Sulphate reduction involves the consumption of a substantial amount of hydrogen ions and the production of HS- at certain pH levels (Hassen, Hamzaoui-Azaza, and Bouhlila, 2016; Santucci, Carol, and Kruse, 2016; Werner et al., 2013). Further evidence to the previous observations are the calculations of the saturation index for calcite (SIcc) and its CO2 pressures, figure 1.5-2b&c. At common temperatures and pressures, the dissolution of calcium sulphate (CaSO4) is in equilibrium with the solid phase of gypsum, but not with anhydrite. There is no natural source of gypsum present in the Semarang lowlands (Purnama and Marfai, 2012; Putranto and Rüde 2016; Rahmawati and Marfai 2013), it is understood that there is an additional source of sulphate, as alkalinity and pH showed highly variable concentrations. Thus, these are thought to be point-source pollution from urban and industrial wastewaters. Dug well owners know that their groundwater has poor quality by its smell and from common knowledge. Either way there are only a few cases where dug well owners preferred groundwater (Artesian wells) or PDAM water over bottled water.

Conclusions and Future research

Semarang city is currently developing, and some of the new pressures appear to be influencing the groundwater quality of the city. This has been viewed throughout the results of this thesis, and therefore of the study area of Semarang lowlands, in the North of the city. In spite of the anthropogenic pressure on the groundwater of Semarang lowlands, there is a natural conditioning factor which defined the groundwater of the unconfined aquifer. This natural conditioning has been unveiled with the hydrogeochemical analysis in this thesis, where the compounds of sodium, calcium, bicarbonate and chloride were identified as the characterising hydro-geology of Semarang lowlands. The local geology along with industrial activities are not the only influence to the local groundwater, also two kinds of pollution seem to be playing a role in the groundwater chemistry of Semarang lowlands. The first one is diffuse, related to the intrusion of sea water into the aquifers, and the second kind is point-source, linked to the poor-quality sanitation infrastructure of Semarang lowlands. The first, sea-water origins were linked to industrial abstractions of groundwater from deeper parts of the aquifer. The point source pollution was found through the presence of E. coli throughout all the dug well samples, nitrite, and some nitrate. Along with the statistical analysis backing to these findings. Therefore, seawater intrusion and sewage were identified as sources of pollution to the unconfined aquifer, also known as Alluvium aquifer.

In this research a great number of evidences of the influence of the ongoing anthropogenic activities happening in Semarang lowlands was found. However, this does not assure these findings to be definite. Other processes not covered in this research cannot be discarded to have an impact to groundwater. These include volcanic emissions, and climate change. It would be of great improvement to carry out a continuous monitoring of the shallow aquifer of Semarang lowlands.

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