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Origin and distribution of nitrate in water well of settlement areas in Yogyakarta, Indonesia


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Abstract Pollution of nitrate in water wells in Yogyakarta City was reported to increase for two decades. This study aimed to describe nitrate contamination in the water wells of colorectal cancer (CRC) and inflammatory bowel disease (IBD) patients, previously described elsewhere. Nitrate and chloride content of 150 water samples from the wells of patients with CRC and IBD who were residing in Yogyakarta, Sleman, or Bantul districts were examined. Description of nitrate contamination was presented in the form of box plot charts and map. Kruskal-Wallis analysis was used to measure the difference of nitrate concentration in three areas of study. Comparisons of nitrate and chloride concentrations were used to determine the source of nitrate contamination in water well. Fisher's exact test was used to

describe the relationship of well distance with the septic tank to nitrate concentration in water well. The wells in Yogyakarta City had the highest median nitrate content compared to Sleman and Bantul ($P = 0.001$) with the median of 56.6, 13.1, and 7.7 for Yogyakarta, Sleman, and Bantul, respectively, and most tested samples exceed WHO safe drinking water standards. The spread of nitrate contamination has occurred in areas adjacent to Yogyakarta City compared to the previous report. The ratio of nitrate to chloride (1–8:1) suggested that the source of nitrate contamination in water wells in the study area came from feces due to inadequate on-site sanitation. The mapping showed nitrate contamination in water wells in Yogyakarta City, Sleman, and Bantul districts had spread according to urban development.

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Introduction

The development of Yogyakarta City has grown rapidly and passed the administrative boundaries of the city so that it covers parts of Sleman and Bantul districts, which known as urban agglomeration areas of Yogyakarta (Pemerintah Provinsi DIY 2005). Yogyakarta City and its agglomeration region have grown into a crowded city, thus, potentially health problems may arise. Some problems faced by the government and communities are the decreasing quality of clean water and improper urbanization process including improper application of

on-site sanitation systems as Yogyakarta City overlying a shallow unconfined groundwater condition. Since the 1990s, researchers reported the decreasing quality of water wells in Yogyakarta and surrounding settlement areas, due to improper sanitation (Heng et al. 2010; Putra 2010).

High concentrations of nitrate which exceeded the safety level for consumption were detected and reported to be in the water wells of Yogyakarta (Heng et al. 2010; Putra 2010). Consumption of high nitrate in drinking water continuously can have a negative impact on health. Several epidemiological investigations reported an association between consuming high nitrate in drinking water and human health such as rectal cancer (Chang et al. 2010; Kuo et al. 2007), colon cancer (De Roos et al. 2003), proximal colon cancer (McElroy et al. 2008), colorectal cancer (CRC) (Espejo-herrera et al. 2015), gastric cancer and prostate cancer (Morales-suarez-varela et al. 1995), thyroid cancer (Ward et al. 2010), childhood brain tumor (Weng et al. 2011), non-Hodgkin lymphoma (Ward et al. 1996), insulin-dependent diabetes mellitus among children (Kostraba et al. 1992), and methemoglobinemia among infants and children (Sadeq et al. 2008). We recently reported the correlation of high nitrate in water well and CRC (Fathmawati et al. 2017). Around 70% of people living in the Yogyakarta Special Province rely on water wells as a source of clean water; this dependency on polluted sources may increase the risk posed by the pollution of nitrates in the water well.

Sudarmadji reported that the average nitrate in water wells in Yogyakarta Municipality in 1985 was about 2.8 mg/L (Sudarmadji 1991). Another study in Kotagede Sub-district, Yogyakarta, which was conducted by Smith et al. in 1994–1996 reported that nitrate in the water well in this area was three times higher than the World Health Organization (WHO)'s guideline for drinking water (Smith et al. 1999). Putra (2007) stated that the concentration of nitrate in water wells in Yogyakarta City tended to increase rapidly over the two decades since 1985 (Putra 2007). The latest studies reported that high nitrate concentrations in water wells were also found in Minggir Sub-district, Sleman District (Oudone 2014), Gamping Sub-district, Sleman District (Wiguna 2014), Bantul Sub-district and Bambanglipuro Sub-district, Bantul

District (Prastoro 2009) as the agglomeration areas of Yogyakarta City.

This study described the increased levels of nitrate contamination in water wells of three municipalities of Yogyakarta Special Province, along with comparisons with previous studies providing conclusive evidence of unhealthy changes of nitrate contamination within three decades which exceed the WHO recommendations for safe and sanitary water requirements.

Material and methods

Study area and sample collection

This study was located in Yogyakarta City and two adjacent districts, Sleman and Bantul. A total of 150 samples were taken from the water well of the residences of patients with CRC and inflammatory bowel disease (IBD) who were treated in Dr. Sardjito General Hospital. Patient selection was based on case and control criteria as has been disclosed in the previous study (Fathmawati et al. 2017). Water sampling was conducted by trained personnel in accordance with the procedures for sampling water well issued by the National Standardization Agency (SNI 6989.58:2008). Samples were taken in wet season (February to March 2016). Water parameters examined consisted of the concentrations of nitrate and chloride. The concentrations of nitrate and chloride were analyzed in the Water Chemical Laboratory of Major Center of Environmental Health Engineering and Disease Control (MCEHE-DC) of Yogyakarta using American Public Health Association (APHA) Standard 2012, section 4500-NO₃ for testing nitrate levels and using SNI 06-6989.19-2009 (Badan Standardisasi Nasional 2009) by argentometric methods for testing chloride levels.

Field data sheets were used to record the observations of the condition of the wells which included the identity of the owner of the well, location, depth, age of the well, and the distance of well from any potential source of contaminants (septic tank).

Methods for estimating the source of nitrate contamination

The ratio between nitrate and chloride concentration was used to analyze whether feces was the main source

of nitrate contamination in water well (ARGOSS 2001; Morris et al. 2003).

Statistical analysis

Kruskal-Wallis analysis was employed to show the difference of nitrate concentration in three areas of study. Fisher's exact test was used to describe the relationship between nitrate in water wells and the distance of the wells from potential sources of pollution. For this test, nitrate was divided into two categories based on the WHO guidelines for drinking water (≤ 50 and > 50 mg/L) (WHO 2011). The distance of well from the septic tank was also divided into two categories (≤ 10 and > 10 m). Stata version 12 was used to conduct the statistical analysis and the box plot chart. Two-sided P value ≤ 0.05 was considered as statistically significant. ArcMap version 10.2 was used to construct the map.

Results and discussion

Results

Samples in this study were taken from three areas in Yogyakarta Province, including the city of Yogyakarta (14 of 14 sub-districts), Sleman (16 of 17 sub-districts), and Bantul (13 of 17 sub-districts). Figure 1 shows the location of the study and comparison of the location of this study with previous studies. The research area of Sudarmadji (1991) was located in Yogyakarta City. Putra (2010) expanded the research on nitrate contamination in Yogyakarta City and its agglomeration. This recent study covered almost all areas in Yogyakarta City, Sleman District, and Bantul District. Two sub-districts in Bantul were not included, since the population did not use water well as a source of water, while two others and one sub-district in Sleman did not meet the subject criteria as described in the previous study (Fathmawati et al. 2017).

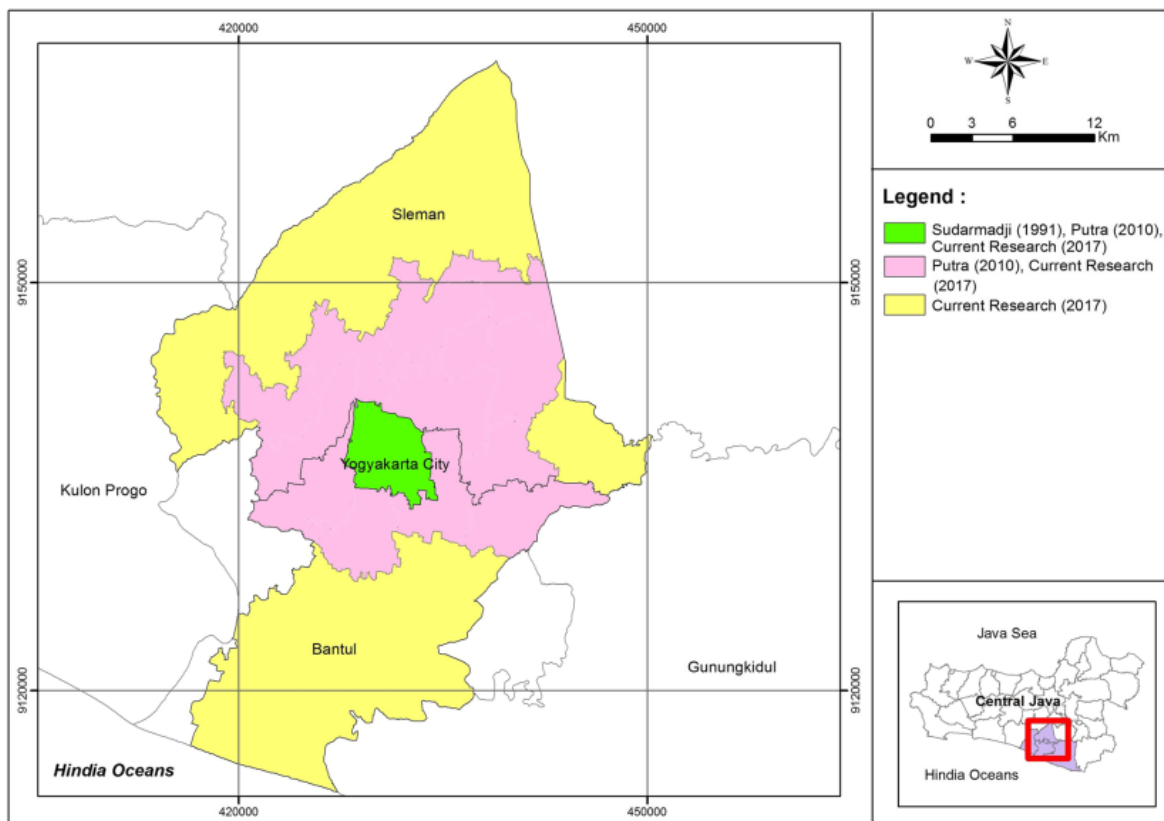


Fig. 1 Map of research area in Yogyakarta Special Province, comparing the coverage of research area among three different studies (see legend) spanning over 30 years

Characteristics of study samples can be seen in Table 1. Most of the samples (54.7%) came from Sleman. Samples were taken from wells which are mostly located in the urban area (93.7%). Many of the wells (43%) were less than 36 years old. Half of the wells (50%) observed had the depth of less than 10 m. Most of the wells (76%) were less than 10 m from septic tanks and 18% of wells contained nitrate more than 50 mg/L.

The wells observed in this study were dug wells which abstract water from shallow unconfined aquifer. Figure 2 shows that the wells in the study site were shallow with a maximum groundwater depth of 16 m b.g.l. (below ground level). According to Hendrayana and de Sousa Vicente (2013), the regional groundwater flow on the study area is from north to south with the slope of water table that gradually diminishes, change of groundwater flow pattern occurs in area adjacent to rivers, in which most of the local groundwater flows (Fig. 3). It should be bear on mind that most of the community in this region are depending on dug wells for their daily life, that is on each house, there is a dug

well and a septic tank system. On the crowded area, it may happen that one dug wells may be affected by many septic systems. Thus on this study, the nearest septic systems is assumed to be the most influence to the nitrate content in the water well.

Figure 4 shows the distribution of nitrate in three areas. A significant difference of nitrate concentration was detected between the three areas of study (P value = 0.0001). Yogyakarta City had the highest median of nitrate in water wells compared to Bantul and Sleman districts. The median nitrate in water well in Yogyakarta was 56.6 mg/L NO_3^- (min. 11.25, max. 116.01), Sleman District was 13.1 mg/L NO_3^- (min. 0.25, max. 65.62), and Bantul District was 7.7 mg/L NO_3^- (min. 0.04, max. 108.29). In Bantul, six wells had nitrate concentrations far above the median.

Figure 5 shows the distribution of nitrate contamination in the study area. The wells in Sleman and Bantul districts bordering with Yogyakarta City had high levels of nitrate (>50 mg/L). Wells with nitrate content of 10.1–50 mg/L were found to spread in the northern and southern regions of the study area.

Figure 6 shows the ratio of nitrate and chloride in the study area. Most observed wells in the Bantul District had the ratio of nitrate and chloride above the 1:20 line. All research areas had wells with nitrate/chloride ratio of 1–8:1 indicating possible fecal contamination.

When comparing nitrate concentration and distance of water well with septic tank, we found a significant correlation between distances of well to septic tank with nitrate concentration in water well (P value = 0.018) as shown in Table 2.

Discussion

Observation of the increase of nitrate pollution in water wells in Yogyakarta needs to be done to avoid more serious pollution which may ultimately impact on public health. The results of research conducted 10 years ago showed a tendency of increased nitrate in water wells, especially in the urban areas of Yogyakarta. This study compared the condition of three observation areas for nitrates in water well with two previous studies conducted by Sudarnadji (1991) and Putra (2007). Although this current study used different sampling methods from the previous two studies, the results of this study provide conclusive evidence concerning the condition of water wells in the area of Yogyakarta and its surroundings within 30 years. Sudarnadji (1991) observed 293

Table 1 Sample characteristics

Characteristics	Frequency	Percent
Source of samples		
Yogyakarta City	24	16.0
Sleman District	82	54.7
Bantul District	44	29.3
Urban/rural classification		
Urban	140	93.3
Rural	10	6.7
The age of well		
≤36 years	65	43.3
>36 years	44	29.3
N/A	41	27.3
The depth of well		
≤10 m	75	50.0
>10 m	50	33.3
N/A	25	16.7
The distance of well to septic tank		
>10 m	36	24.0
≤10 m	114	76.0
Nitrate concentration in the wells		
≤50 mg/L	123	82.0
>50 mg/L	27	18.0

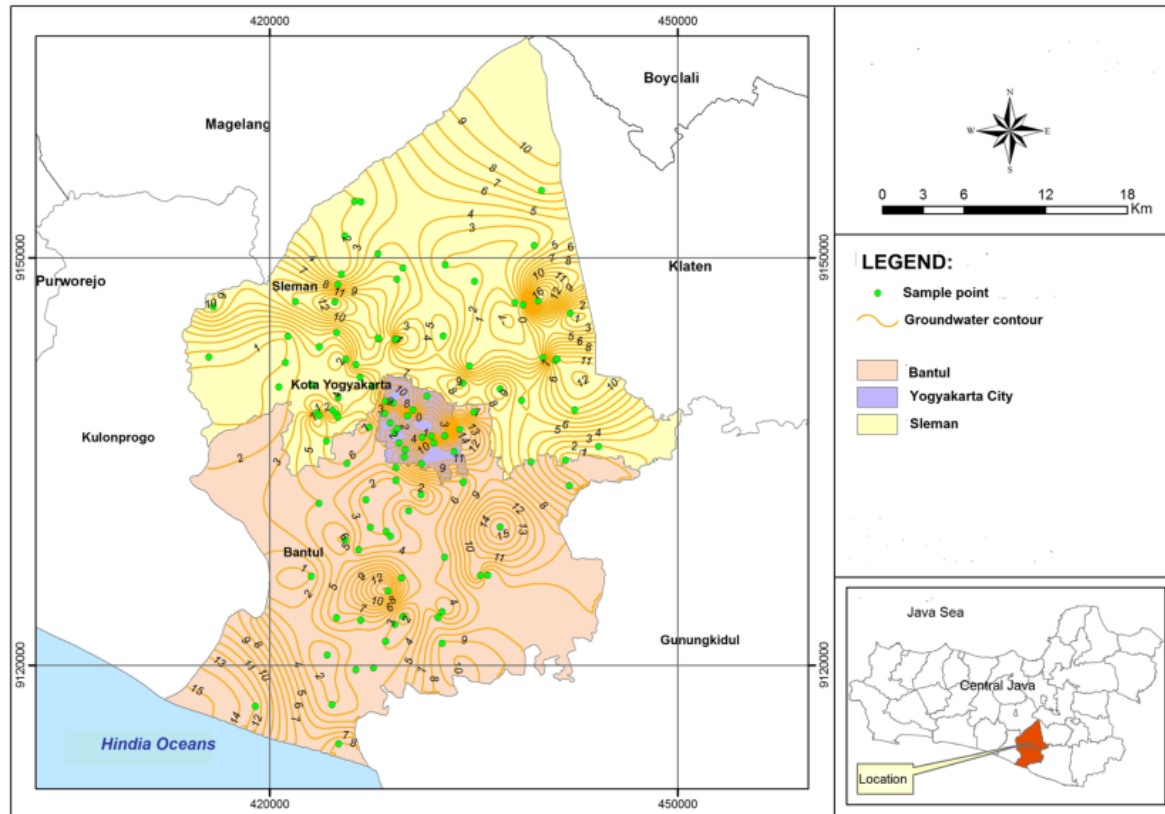


Fig. 2 Map of groundwater depth below ground level

wells and Putra (2007) observed 144 wells, both used random sampling to determine the well observed (Putra 2007; Sudarmadji 1991). This current study observed 150 wells by case-control methods (Fathmawati et al. 2017) and covered a wider area than the previous studies (Fig. 1).

Nitrate in water wells in Yogyakarta City

Sudarmadji (1991) reported that in 1985 nitrate was not a problem in water wells in Yogyakarta Municipality (Sudarmadji 1991). Twenty years later, the concentration of nitrate increased, especially in urban areas. An increasing of nitrate concentration in water wells was found in Yogyakarta and its agglomeration which became more than 10-fold since reported by Sudarmadji (Putra 2007). The current study reported that the median of the concentration of nitrate in water wells in Yogyakarta City increased almost 20 times compared to the results obtained by Sudarmadji (1991) and almost twice compared to the results obtained by Putra (2007),

consecutively 2.8 and 31.5 mg/L. This current study in 2017 showed that the median of nitrate in water wells in Yogyakarta City was 56.6 mg/L (Fig. 4).

This study shows a similar trend to the results obtained by Putra (2007) which revealed an increase in nitrate concentrations in water wells in Yogyakarta City over time. Studies conducted by several researchers indicated that an increase of nitrate is correlated with the development of urban areas in Yogyakarta which was not followed by adequate sanitation infrastructure (Putra 2007, 2011; Heng et al. 2010; Wiguna 2014; Prastoro 2009).

Nitrate in water wells in Sleman and Bantul districts

The wells in the areas adjacent to Yogyakarta City, namely Sleman and Bantul districts, began to show the occurrence of nitrate contamination with high concentration. Although Bantul had the lowest median nitrate concentrations, several villages which were located adjacent to the City of Yogyakarta had nitrate

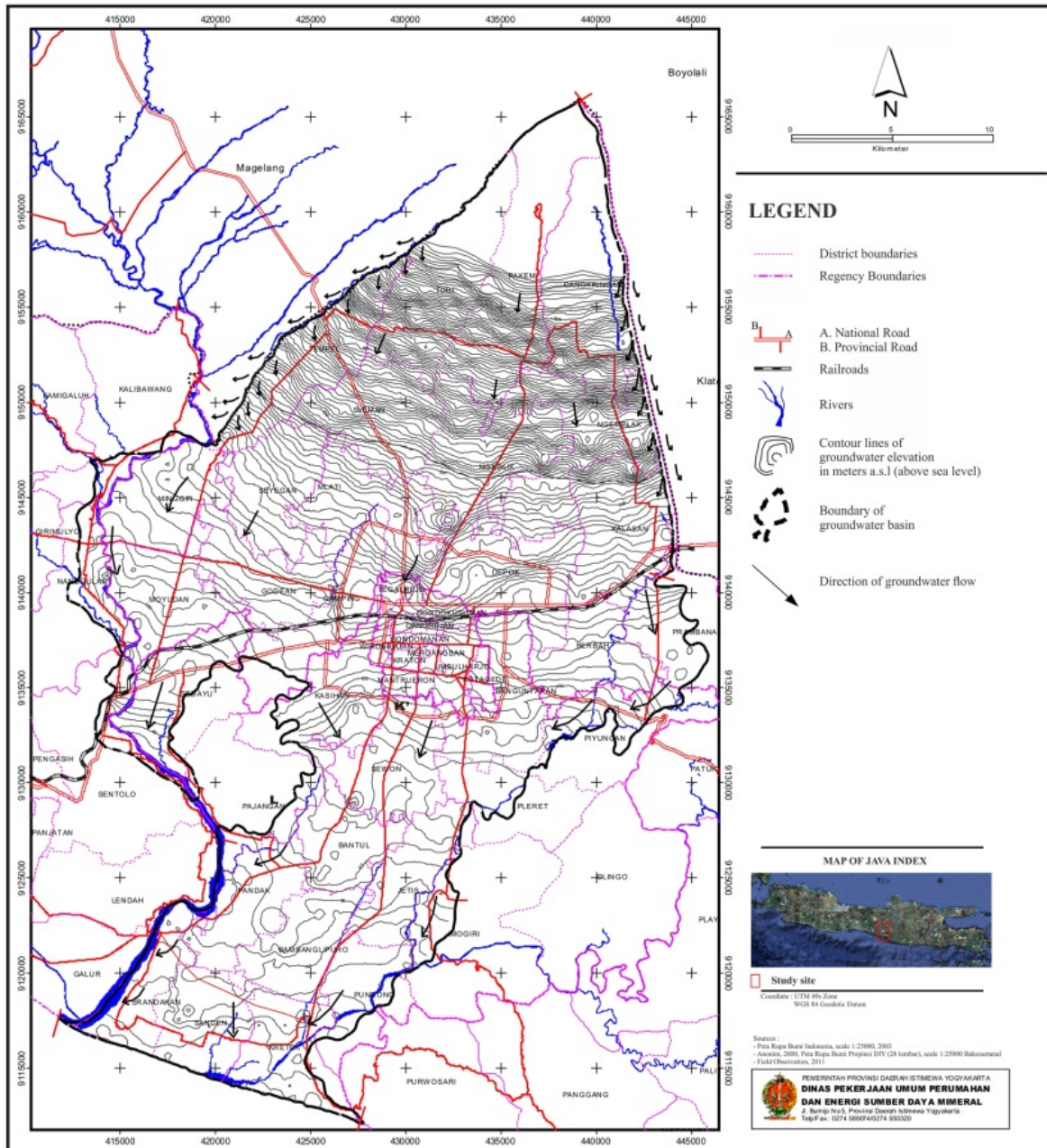


Fig. 3 Yogyakarta-Sleman groundwater basin map (after Hendrayana and de Sousa Vicente 2013)

concentrations exceeding 50 mg/L NO_3^- . As can be seen in Fig. 5, in Bantul there were six wells having nitrate exceeding WHO standards for drinking water and had extreme concentrations. These six areas as shown in Fig. 5 were in the areas adjacent to Yogyakarta

City (see the red and orange marker). This finding was confirmed by the previous study. Studies in Bantul and Bambanglipuro sub-districts in the year 2009 showed that the nitrate concentrations almost reached the maximum permitted (Prastoro 2009).

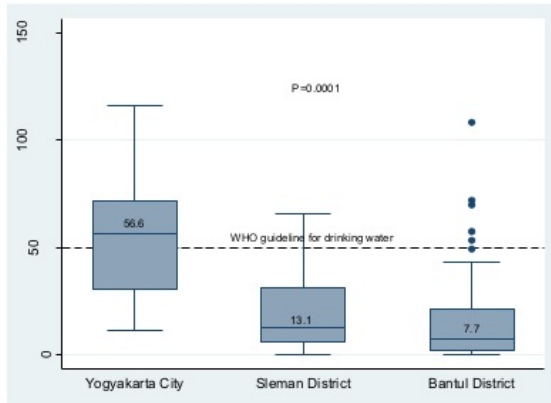


Fig. 4 Distribution of nitrate concentration in study area of the Yogyakarta Special Province. Data showed that Yogyakarta City had the median of nitrate above WHO guidelines (50 mg/L) much higher compared to Sleman and Bantul districts

Sleman District had median concentrations of nitrate below the WHO standard for drinking water, similar to Bantul District. However, this study found eight wells

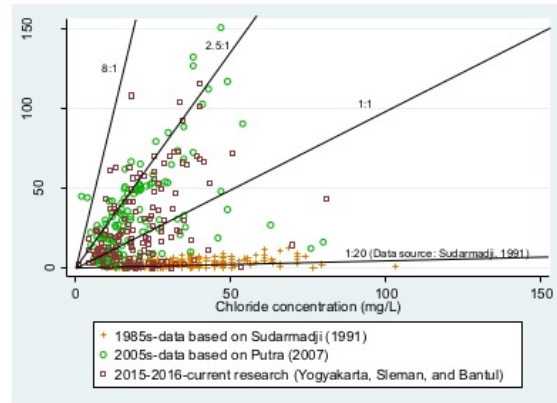


Fig. 6 Nitrate-chloride concentration ratio on well water in the study area, comparing this report with Sudarmadji (1991) and Putra (2007). Overall the nitrate/chloride ratio (1–8:1) indicates nitrate may originate from feces

with nitrate concentrations exceeding the standard (see the orange marker in Fig. 5). This finding was supported by previous studies which obtained well samples in two

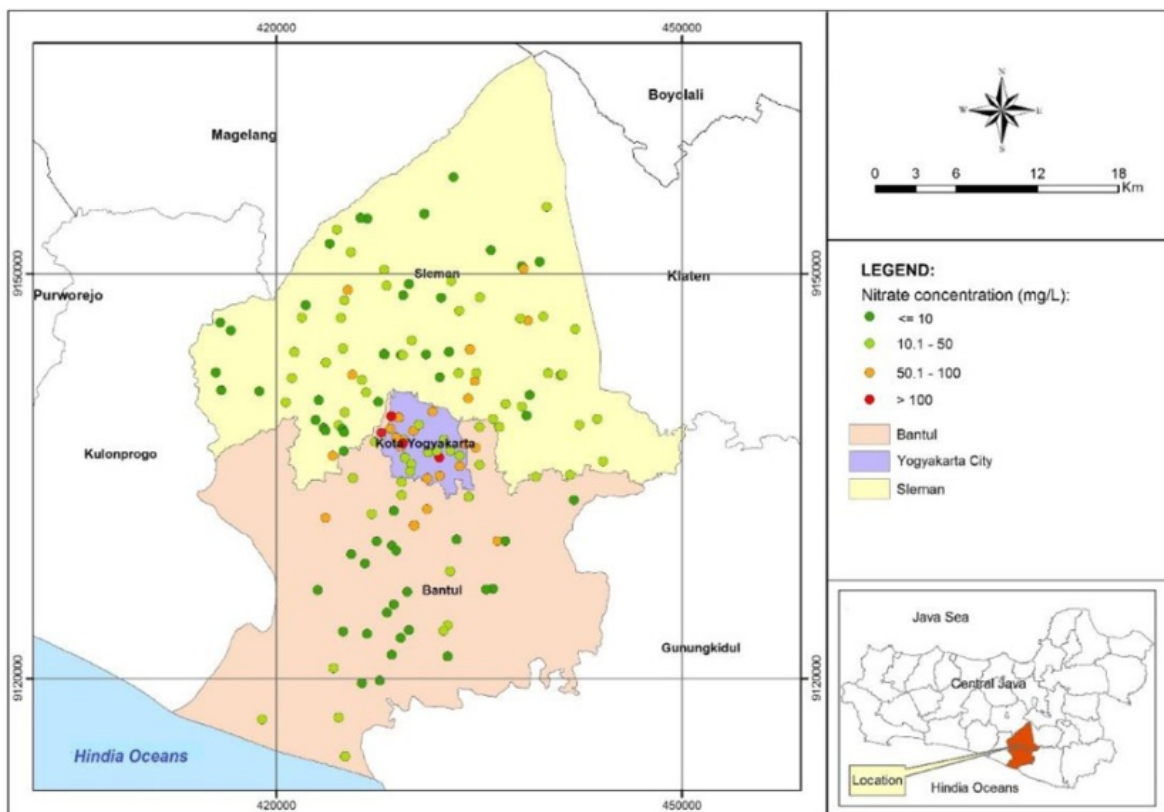


Fig. 5 Map of nitrate concentration in sampled wells in research area

Table 2 Nitrate concentration and the distance of wells to septic tanks

Nitrate concentration	The distance of wells to septic tank				Total		P value*
	> 10 m		≤ 10 m		n	%	
	n	%	n	%			
≤ 50 mg/L	34	27.6	89	72.4	123	24.0	0.018
> 50 mg/L	2	7.4	25	92.6	27	76.0	
Total	123	100.0	27	100.0	150	100.0	

*Fisher's exact test, with two tailed P value ≤ 0.05 considered as statistically significant

regions located in the agglomeration region having nitrate exceeding the WHO standard. In 2014 observation, nitrate concentrations which exceeded twice the permitted concentration were found in Gamping Sub-district, Sleman (Wiguna 2014). In Minggir Sub-district, Sleman, the nitrate concentration was also exceeding the maximum permitted (Oudone 2014).

The distribution of pollution was related to the development of the two districts which were the agglomeration areas of Yogyakarta City. The population growth of Yogyakarta Province from 2000 to 2010 was 1.04% per year and the projected population growth from 2010 to 2015 increased to 1.19% per year. Although the overall growth increase was only 0.15, it still has an impact on socio-economic life because it involved the population increase of more than 40 thousand people per year (Bappeda D.I. Yogyakarta 2016). Additionally, the cities of Yogyakarta, Sleman, and Bantul were the places which have the greatest number of colleges. As a result, the concentration of settlements was in these three areas.

Source of nitrate contamination

Nitrate/chloride ratios in this study were compared with data obtained by Sudarmadji in 1985 (Sudarmadji 1991). The ratios of nitrate/chloride processed from Sudarmadji (1991) were not only used to predict the source of nitrate contamination in water well but also to prove the increase of nitrate concentration in the settlement areas. This method has also been used by Putra (2007) in his analysis. Most of the wells which were observed in this study showed ratios of nitrate/chloride between 1 to 8:1. ARGOSS (2001) stated that the ratio of nitrogen and chloride in feces were 2–2.5:1. Under aerobic conditions, nitrogen will be oxidized to nitrate.

Therefore, the ratio of nitrate and chloride concentrations can be used to prove that the source of nitrate contamination in water well came from feces (ARGOSS 2001). Morris et al. (1994), however, stated that if another wastewater came into the disposal system (e.g., gray water), the ratio was no longer valid because it may contain ancillary chloride. Therefore, based on ARGOSS (2001) experience, if the ratio of nitrate and chloride concentrations in water well ranges from 1 to 8:1, nitrate may be originated from feces.

This result may infer that the source of nitrate contamination came from feces. Although there were wells which have the ratio of nitrate/chloride 1:1–20, it can be estimated that feces were the source of nitrate contamination (Fig. 6). This finding was in accordance with Putra (2007) who considered the condition of the environment of wells in the urban area. His observation showed no other source of pollution but the septic tank. The data which were obtained in this study supported the conclusion that feces were a source of nitrate pollutants because the majority of the wells had a distance from septic tanks less than 10 m (Table 1).

In addition, Fig. 6 also showed an increased level of nitrate gradually in water wells from 1985 study. This increase occurred due to the persistence of nitrate and the increased rate of nitrate leaching from time to time (Putra 2007). This condition was in accordance with ARGOSS (2001) statement that high nitrate contaminated groundwater not decrease rapidly even though the nitrate load was reduced or eliminated. The city of Yogyakarta and the surrounding urban areas had a population density of about 12 thousand inhabitants per square km (Bappeda D.I. Yogyakarta 2016). Meanwhile, domestic waste water treatment was generally involving inadequate on-site sanitation systems. Centralized wastewater treatment presently can only serve

about 20% of houses in Yogyakarta City. This condition causes high levels of nitrate to remain in the groundwater which tends to increase over time.

Nitrate pollution in water well by human excreta can also be correlated with the distance of wells to septic tanks with less than 10 m as shown in Table 1. A total of 114 of 150 (76%) wells tested had a distance less than 10 m to the septic tank. The concentration of nitrate in water wells was significantly different if it was seen by the distance of wells to septic tanks. We found 92.6% of wells containing nitrate more than 50 mg/L had a distance to the septic tank less than 10 m (Table 2). Although a distance of 10 m is not the limit of nitrate transport from the sources of pollution to the wells, the finding provides conclusive evidence that the source of nitrate pollutants was within close proximity to the wells.

This analysis was also supported by Smith et al. (1999) who found that the sediments of urban areas in Yogyakarta, Bantul, and Sleman were consisted of coarse sand deposit. Thus within this region, the distribution of nitrate concentrations in the groundwater was found higher closer to the septic tank due to high permeability of the aquifer. As mentioned by Putra and Indrawan (2014), the permeability of aquifer in this region ranged from 3 to 700 m/day. Regarding the source of nitrate from agricultural source, it should be noted that the agriculture area in this region was a paddy/rice field area. Putra (2007) has reported that water well under paddy/rice field were much lower than on the dwelling area in the average of 16.5 mg/L, this value was in agreement with the value of anticipated groundwater nitrate concentrations for paddy/rice field sited on shallow groundwater system by BGS (2004). This further supports that inadequate sanitation system from dwelling area was potential source of nitrate.

Nitrate reduction can take place due to the denitrification process but it is not effective in water well so that the nitrate behavior is almost the same as conservative substances. In other words, nitrate is a stable ion, which does not easily change physically, chemically, or biologically in its journey toward ground water (Freeze and Cherry 1979), which means that if it has entered the water well and there is a close source of nitrate, the concentration will not decreased but will always increased in water well. Meanwhile Corniello et al. (2007) added that nitrate reduction may take place through the progressive dilution effect of water percolation. However, the decreased nitrate concentrations in water well will not take place if the disposal of domestic

waste, especially human excreta, is not performed adequately and at a safe distance from the water source. ARGOSS (2001) stated that although dilution could reduce the nitrate content in groundwater, it will not be effective if the nitrate load was high and came from a large polluting source within a large area.

Based on this report, the risk of increasing levels of nitrate contamination may further increased in Yogyakarta if an integrated and planned wastewater management program is not implemented soon (Prastoro 2009). Therefore, the integrated waste management is absolutely needed to protect both water well from contamination and sources of drinking water for people who are in danger of nitrate exposure.

Communities should not consume water wells which have been contaminated by high nitrate levels considering the negative health effects it causes (Daniels and Mesner 2010). Information about this risk should be communicated to the community by the local government through the local health service. Monitoring the water wells quality of the population should be done periodically by the local health service through the community health center. This preventive approach should be done to ensure the safety of water consumed by the community.

A limitation in this study was the method of determining the source of nitrate contamination using the ratio of nitrate and chloride. This method had a limitation because it did not consider other sources of nitrate contamination. ARGOSS (2001) stated that to calculate the relative contribution of each source of nitrate contamination was difficult. Eby (2016) recommended to use nitrogen isotope analysis ($\delta^{15}\text{N}$) to ensure the source of nitrate contamination in water wells.

Conclusions

Increasing concentrations of nitrate in water wells have been occurring for the last 30 years due to inadequate on-site sanitation. Integrated wastewater management can prevent nitrate contamination by human excreta into the water wells and protect residents in and around Yogyakarta from potential health risks associated with drinking water with high levels of nitrate.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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