Groundwater Contamination Due to Nitrate in a Coastal Aquifer: Conjecture and Identification Along the West Coast from Katana to Negombo, Sri Lanka

P. Ratha, K. D. W. Nandalal, S. B. Weerakoon
Department of Civil Engineering
University of Peradeniya
Peradeniya, Sri Lanka

H. A. Dharmagunawardhane, H. M. T. G. A. Pitawala
Department of Geology
University of Peradeniya
Peradeniya, Sri Lanka

Abstract—Higher amount of nitrate in drinking water can cause severe methaemoglobinemia and cancer in infants and adults. Land-uses and anthropogenic activities accompanied by hydrogeological conditions cause nitrate contamination in groundwater. This study aimed at estimating and identifying using box-plot, empirical approach and nitrate:chloride ratio (1:1 – 8:1) as feasible methods to prove the possible sources in the western coastal area in Sri Lanka from Katana to Negombo. The outputs show that a high nitrate in groundwater of the area is originated mainly from on-site sanitations (i.e. soakage, pit latrine, septic tank) under denser populated settlements, while agriculture plays as a secondary source. Current groundwater nitrate levels from analyzed water wells indicated that about 30% of the water samples exceed the Maximum allowable value of 10 mg/L NO₃⁻, however, samples with nitrate greater than 50 mg/L were not found. Denitrification processes may also contribute regulate nitrate levels in the area. Although nitrate concentration at present is not alarmingly high in the area, an increasing trend of contamination is expected with population, settlements and agriculture growth. Therefore, appropriate management and special care from relevant parties should be set up to rectify the problems and to use precious groundwater resource sustainably.

Keywords—Nitrate contamination, coastal aquifers, Sri Lanka, methaemoglobinemia, nitrate:chloride ratio, on-site sanitations

I. INTRODUCTION

Coastal groundwater has become a precious resource for man in many parts of the world, where surface water is scarce. Of global use, groundwater consumption is mainly for domestic, agricultural, and industrial purposes. Groundwater needs to be free from contamination; however, pollution of groundwater is a main issue worldwide, which affects human health and economy. The restoration and remediation are not easy and may be impractical, if contaminated. Rapid urbanization, anthropogenic stresses, and climate change have been witnessing an increased pressure on groundwater pollution globally by posing diversity of threats to groundwater and consequently depleting both quality and quantity [1]. Over 30 years ago, nitrate, chloride, bacteria, and pesticide detections have been increasingly encountered. Among these pollutants, groundwater nitrate contamination has been already globally recognized as a growing problem [2] in Europe, Asia, south Asia, and Africa. Nitrate (NO₃⁻) is a main pollutant commonly found in groundwater with maximum standard limit of 50 mg/L (or 10 mg/L under natural condition as NO₃⁻ – N) and of 3 mg/L for nitrite (as NO₂⁻) [3].

Nitrate contamination in groundwater is derived from different sources and processes. The origins of nitrate contamination can be natural and anthropogenic associated with four main means such as (i) human and animal waste accumulation and decomposition (point source), (ii) nitrogen fertilizer application in dense agriculture (non-point or diffuse source), (iii) earth organic matter decomposition, (iv) natural nitrate occurrence in desert area with addition of nitrate from atmosphere through rainfall [4,5]. Under aerobic conditions, high nitrate can be expected from a significant percentage of organic nitrogen, which will be oxidized to form nitrate (nitrification) based on biological conversion of ammonium (NH₄⁺) through nitrite. Nitrate (also chloride) is considered as very mobile, conservative and often stable (thus persistent) contaminant in groundwater as less adsorbed or retarded in the soil and is thus easily leached by heavy rainfall and infiltrating water [6]. Poor fertilizer application, surface spreading of animal manure, and improper on-site sanitation systems are the greatest sources of high nitrate content in groundwater. Moreover, population density, types of settlements, rainfall recharge rate, sanitation behaviors, and per capita water use highly contribute to nitrate – nitrogen contamination load. Under shallow groundwater and underlain by thin permeable soils and a permeable aquifer, increase in widespread nitrate contamination exists to deteriorate groundwater quality [7]. Using elevated nitrate content in drinking water can commonly cause public health risk and environmental pollution. It acutely causes health effect of methaemoglobinemia or infantile cyanosis known as “blue baby syndrome”, in which blood lacks the ability to carry adequate oxygen to the individual body cells. Infants below three months are at the most risk. In addition, long term exposure (chronological) to excessive nitrate amount has been found in many literatures to be a risk cancers for older people (i.e. gastric, colorectal, bladder, urogenital and brain tumor), esophageal and stomach cancer, diabetes, and thyroid.
The inland area from Katana to Negombo is characterized by sands on a prior old beach plain formation underlain by interbedded clay followed by the crystalline bed rock granite and pink granite. Small portion of laterite is found in the central west close to Negombo lagoon, from Kochchikade through Negombo until Kandana (Negombo-Katunayake – Seeduwa – Ja-Ela – Kandana) [16]. Available borehole data from Water Resources Board (WRB) represents that the geological succession of the area comprises of regosols and latosols of sands with significant clay (as alluvium soils) about 12–24 m thick resting on the hard rock. Comparatively lower thickness is found in central eastern parts while deeper thickness in the central, western and southern parts of the area. Additionally, the sand layer thickness seems to have high variation by varying from about 2–20 m underlain by interbedded clay layer varying from about 5–19 m. The Geological formation of the area has been hydrogeologically categorized as shallow (unconfined) aquifers and moderately deep aquifers (semi-confined). The main aquifer consists of sand and sandy clay with thickness ranging from 10 to 15 m, underlain by another aquifer whose thickness ranges between 8 to 15 m [17]. The unconsolidated sand belt running along the coastal belt particularly from Ja-Ela, to Seeduwa and Katunayake up to Negombo. The water level is estimated to be around 5–8 m below ground level (bgl) with flow direction from north to south and east to west and then discharge into Negombo lagoon at the west (see Fig. 1).

III. METHODOLOGY

A. Water sample collection

A total of 26 groundwater samples were collected during July to August 2018 from evenly spaced locations of dug and tube wells based on land-uses of the study area. The samples taken were bottled in washed pre-cleaned unfiltered polyethylene (HDPE) containers with inner caps without leaving air space to avoid internal evaporation [18]. It is well noted that the sampling was considered as only one-time sampling. The water from tube wells was sampled after 5-minutes purging out to remove any stagnant oxidized water in the well casing. Water parameters examined in this study contained nitrate and chloride concentrations. The contents of nitrate and chloride were analyzed using standard methods, in the Laboratory of Environment Department of Civil Engineering, University of Peradeniya, Sri Lanka. Analytical instrument HACH/DR2010 Spectrophotometer was used to determined nitrate concentrations, while Titration method was operated for analyzing chloride amounts using Potassium Chromate indicator titrated against Silver Nitrate solution. Physical parameters like pH, electrical conductivity (EC) and temperature were measured in the field with sampling time. The recorded information like the observations of the well conditions (i.e. types of well, location, depth, usages) and land-uses was done by using field data sheets.

B. Estimating nitrate amount and contamination sources

Believed to be a very conservative ion in groundwater, nitrate and chloride contaminants have been chosen as an indicator contaminant for the evaluation nitrate contamination sources. The concentration of both contaminants in groundwater recharge can be semi-quantitatively estimated...
utilizing following empirical method in (1) suggested by ARGOSS [6]; Foster & Hirata [19].

\[
C_{\text{N/Cl}} = \frac{1000 \times a \times P_d \times F}{(0.365 \times P_d \times U_{\text{non}}) + 10 \times I}
\]  

(1)

Where,

- \(C_{\text{N/Cl}}\): The concentration of the nitrate or chloride in recharge [mg/L]
- \(a\): Unit weight of nitrogen or chloride in excreta [kg/cap/a]
- \(P_d\): Population density [cap/ha]
- \(F\): Proportion of excreted nitrogen or chloride leached to groundwater (range 0 – 1.0)
- \(U_{\text{non}}\): Non-consumptive portion of total water use [L/d/cap]
- \(I\): Natural infiltration rate [mm/a]

The ratio relationship approach of nitrate to chloride content proposed by Morris et al. [5]: ARGOSS [20] was employed to identify the sources of nitrate in water well at the area whether or not the main source is from feces or agriculture.

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Fig. 1. a) Location of the study area showing land-use categories, b) groundwater table contours and flow directions, and c) east – west cross section of the area.
IV. RESULTS AND DISCUSSION

A. Estimation of nitrate concentration of the area

The most mobile and stable contaminants are nitrate and chloride, especially from on-site sanitation and agriculture, in form of nitrogen in environments where abundant oxygen is available. A person excretes annual average amount of nitrogen with an estimated value of 4 to 5 kg, and about 2 kg/cap/a for chloride [2,5]. These values respond to (a) parameter used in (1), representing as unit weight of nitrogen in excreta. The proportional nitrogen to be leached to groundwater is related to hydrogeological settings of unsaturated materials (see Table1) with reasonable values from 0.01 – 0.6 (1% – 60%). Thin and permeable soil like sand and gravel gives higher percentage of potential leached nitrogen. However, in fractured sediments, possible leached value is up to 1.0 (approximately 100%) [19 - 22]. All these mentioned leaching fraction is F parameter in (1), denoting as proportion of excreted nitrogen leached to groundwater.

<table>
<thead>
<tr>
<th>Hydrogeological settings</th>
<th>Nitrate fraction likely to be leached</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Unconsolidated sedimentary aquifer</td>
<td></td>
</tr>
<tr>
<td>i) Clay, silt, fine sand</td>
<td>Up to 0.3 that could be very low, particularly shallow water table and clays</td>
</tr>
<tr>
<td>ii) Fine – medium sand</td>
<td>Approximately 0.3</td>
</tr>
<tr>
<td>iii) Medium sands and gravels</td>
<td>0.3 – 0.5</td>
</tr>
<tr>
<td>2. Weathered basement aquifer</td>
<td></td>
</tr>
<tr>
<td>i) Thick weathered layer</td>
<td>Up to 0.3, yet could be very low especially where water table is shallow and weathered material clays</td>
</tr>
<tr>
<td>ii) Thin and/or greatly permeable weathered layer</td>
<td>0.3 – 0.5</td>
</tr>
<tr>
<td>3. Fractured consolidated sedimentary aquifer</td>
<td>Up to 1.0</td>
</tr>
</tbody>
</table>

Using known population density and applying the (1) above with similar average amount of non-consumptive water use (Unon) and variation of I and F, the potential amount of nitrate concentration leached from seepage water from in-situ sanitation can be postulated by conducting experimental figuration in Fig. 2. From this figure, the capability of producing troublesome nitrate content from the case of the area is expected greater than 40 mg/L on high nitrogen leaching condition.

However, in the case of the study area dominated by unsaturated zone materials as medium sand, the reasonable value of leaching fraction thus is supposed to be 0.5 (F = 0.5). As a result, the troublesome average nitrate concentration in groundwater in the area should be less than 20 mg/L. This value may be due to less population density and possible dilution from high rainfall of the area. It is well noted that this estimated nitrate is considered as only from sanitary systems.

B. Current status condition of nitrate in water well

The wells investigated in this study were dug and tube wells which abstract water from unconfined aquifer. The wells in the study site were shallow with a maximum groundwater depth of 15 m and with majority of about 5 m bgl. According to WRB borehole dataset and field measurement in 2018, the regional groundwater flow on the study area is from east to west and north to south then discharge to Negombo lagoon in the west (see Fig.1).

It should also be noted that majority of community in the area are relying on shallow dug wells for their everyday lives, which is on most houses, dug wells and on-site sanitation exist. The populous community is particularly found in Ja-Ela in the south and Seeduwa and Katunayake in central southwest and central part, respectively along Negombo lagoon, where are dominated by sandy aquifer. Recently, the 26 analyzed groundwater samples shows that nitrate concentration of the shallow groundwater within the Katana up to Negombo varies from...
from 2 – 27.5 mg/L with mean value of 8.5 mg/L, while the mean chloride in the water well is 4.7 mg/L (min. 0 and max. 11.7). The pH values range from 5.6 to 7.8 with an average of 7.1 (see Table 2). The majority of the samples show higher nitrate concentration observed from non – agricultural as human settlements rather than agricultural areas like coconuts, especially located in south, central south and central parts of the area. This indicates that nitrate maybe comes from sewage systems or septic tanks. The further notation shows that the nitrate values from tube wells were lower than that from dug wells.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Min.</th>
<th>Mean</th>
<th>CV</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO₃⁻</td>
<td>0</td>
<td>8.5</td>
<td>76</td>
</tr>
<tr>
<td>Cl⁻</td>
<td>0</td>
<td>4.7</td>
<td>11.7</td>
</tr>
<tr>
<td>pH</td>
<td>4.5</td>
<td>7.5</td>
<td>0.7</td>
</tr>
<tr>
<td>Temperature</td>
<td>25</td>
<td>35.8</td>
<td>1.7</td>
</tr>
</tbody>
</table>

The result also indicates that 31% of the water samples are above the human affected value of 10 mg/L NO₃⁻, but there is no samples exceeding 50 mg/L [24]. In natural condition, NO₃⁻ concentrations does not exceed 10 mg/L, so that the beyond 10 mg/L is an indication of anthropogenic pollution, poor sanitation, possible indiscriminate use of higher fertilizers [3].

Fig. 3 shows a box plots of nitrate concentration in shallow groundwater from different land-use types within the study area. This diagram indicates a good relationship between land-use categories and nitrate contents. The mean nitrate content and the range of nitrate distribution are highest in populous settlements classified as non – agriculture. This may be because of nitrate leached from domestic waste. The generalized linear model (GLM) under Poisson regression processed in R Studio shows that the non-agriculture (β=0.78) is the most significant factor (P-value = 0.000012 < 0.05) influencing nitrate level at 95% confidence level. The highest median nitrate concentration was 6.6 mg/L NO₃⁻ (min. 2, max. 27.5) recorded non – agriculture (with coefficient variation, CV of 84%), whereas lower median concentration was 3.7 mg/L NO₃⁻ (min. 2.3, max. 12.4) in coconut + paddy cultivation lands (with CV of 65%) and lowest 3.3 mg/L NO₃⁻ in other areas (free or barren lands). The witness for lower concentration of nitrate in the coconut + paddy lands categorized as agriculture may be due to the low frequency of fertilizer application in this land-use type compared to the settlements supplying human waste (i.e. excreta and urine). Moreover, this current study shows that nitrate has high variation with overall CV of about 90% (10% < CV < 100%) and positive correlation of nitrate with major ions, indicating that groundwater is greatly affected by anthropogenic factors rather than accumulated from the aquifer materials (especially on-site sanitation from dense settlements and also agricultural activities) with additional natural factors like climate and hydrogeological settings [25 - 27].

TABLE 2. PROPORTIONAL PERCENTAGE OF POTENTIAL NITRATE LEACHED FROM ON-SITE SANITATION [20]

C. Nitrate contamination sources

Land-use is related to the human activities, and not only land-use greatly roles for identifying nitrate concentration in groundwater, but also another possibility, such as intrinsic hydrogeological conditions. As sea water has very low nitrate amount of <1 mg/L [28] and no evidence from geological source for nitrate in the area exists, higher nitrate may be mainly contributed from the existing sanitation and waste systems with addition from acts of agriculture.

![Fig. 3. Box plot showing distribution of nitrate concentrations based on land-uses in the area.](http://www.ijert.org)
From many experiments, ARGOSS [20] has concluded that where the nitrate/chloride ratio is between 1:1 and 8:1, it is likely that the nitrate is primarily from a faecal source.

If the single source of nitrate and chloride is actually from domestic effluents, the ratio of these ions in underlying shallow groundwater should be approximately 2 – 2.5:1 (or the ratio of chloride to nitrogen in human waste is about 1:2). With greater ratios of nitrate:chloride and outside range of 1:1 and 8:1, the proportional nitrate derived from other factor, non-faecal sources (as inorganic fertilizers) is likely to be higher, even though some may still be derived from faecal matters.

Therefore, the ratio of nitrate/chloride in the study area has proved that the origin of nitrate contamination in water well, on one hand, is fromfeces. On the other hand, some samples from the land-use setting have the ratio of nitrate/chloride in groundwater out of range 1:1 and 8:1. This proves that not all nitrogen from on-site sanitation of the area leached to groundwater, but the source may be from agricultures with fertilizer application. However, the assumption validity of the ratio may not exist once another wastewater goes into the disposal systems (e.g., gray water), which consists of additive chloride [5,31]. It should also be noted that some samples taken from shallow groundwater under dense settlements (not so far from the country’s airport) surrounded by coconuts along main road close to Negombo lagoon in the west contain high nitrate that is also mainly from faecal source. This evidence may be due to very mobile and stable nitrate moving with groundwater flow in the area from east to west with an estimated hydraulic conductivity 12 m/day. Thus, the observed groundwater nitrate could reflect the effect of upstream activities. Since the growth of nitrate leaching concerned with the mushrooming of population and urbanization process as well as agricultural acts, Putra [2] inferred that higher percentage of nitrate leaching and concentration occur in old developed area (i.e. older unsewered area) than new developed area.

Fig. 4. Composite map of nitrate amount and land-uses within the area.

The assumption of this inference is based on few main phenomena, which include (i) ammonium adsorption capacity of the soil zone may be fully utilized in older systems, (ii) ammonia may be poorly adsorbed as saturated and reducing conditions happen in the soil zone under unsewered sanitations are, and (iii) diminution of denitrification process in aquifer. With rapid shallow groundwater extraction, aerobic condition enhances, and nitrogen will be oxidized to nitrate through nitrite from ammonia, which is call nitrification shown in (2). Nitrification plays as a vital role to produce nitrite (NO\textsubscript{2}⁻) and nitrate (NO\textsubscript{3}⁻). This process takes place naturally and is carried out in two steps [34], in which the first step is nitrification that uses a type of bacteria called nitrosomonas. During the process of nitrification, NH\textsubscript{3} (ammonia) is oxidized into NO\textsubscript{2}⁻ (nitrogen dioxide).

\[
\text{NH}_3 + 1.5\text{O}_2 \rightarrow \text{NO}_2^- + \text{H}^+ + \text{H}_2\text{O}
\]

The second part is called nitration process, which utilizes the enzyme nitride oxidoreductase to complete the nitrification process in (3).

\[
\text{NO}_2^- + 0.5\text{O}_2 \rightarrow \text{NO}_3^-
\]

Once these processes are completed, the possible occurrence of nitrate reduction to nitrogen exists through the progressive dilution effect of water percolation, denitrification process and geological conditions, yet it is not effective in water well, hence the nitrate behavior is almost the same as conservative substances. As mentioned earlier, nitrate as stable ion is not easily transformed physically, biologically or chemically in its travel to groundwater [32]. Natural denitrification is also recognized to happen when shallow aquifer environmental redox potentials are less than 300 mV, and when heterotrophic bacteria in presence of organic carbon existing. Absence of organic carbon enhances other chemical species such as sulphides and minerals containing ferrous iron (Fe\textsuperscript{2+}) to reduce nitrate to nitrogen [35]. This condition causes soil microbes to use NO\textsubscript{3}⁻ ions as the electron acceptor during...
microbial respiration and leads to a process of reduction in which first NO$_3^-$ is denitrified as converted to nitrogenous gas and then ferric iron (Fe$^{3+}$) is reduced to ferrous iron, which is serving as indicator of denitrification process [36]. High reducing condition is found in clayed soil due to lower permeability and secondary iron-bearing phases, which control the input of NO$_3^-$ into groundwater [1,37]. Regarding to the normal pH range of groundwater (pH = 5 – 8), dissolved iron will present as Fe$^{3+}$, since Fe$^{3+}$ is solid and insoluble under these conditions [33]. This fact is reasonable to state that the denitrification condition in the shallow groundwater system of the study area was more appropriate years ago, resulting low nitrate concentrations in groundwater (see Fig. 4). Even though, recent nitrate concentration is still low, it will continue to rise as increase in population, settlements and anthropogenic activities. As estimated, a population density of the area is about 1800 persons/km$^2$ and the growth rate of 1.72%, which is high compared to the country population density of 342 persons/km$^2$ with growth rate of 1.1% (est. 2017). Increase in nitrate trend is evidenced if on-site sanitation and optimization of anthropogenic acts (i.e. fertilizer application) are not properly set up.

Lowered nitrate concentration in water well will not exist if indiscrimination of domestic water disposal, particularly human product widely performs with unsafe distance from water sources. Somehow nitrate in groundwater can be reduce through dilution of percolating water, but effectiveness is not valid if the high nitrate load came from an extensive polluting source within a sizable area [20,31]. Thus, appropriate management and monitoring should be integrated to prevent nitrate exposure danger to water well from contamination sources where drinking water is available for people as well as community. The community is not encouraged to utilize high contaminated nitrate, which can cause negative health impacts. Water containing nitrate cannot be uncovered by tasting (like chloride), and removal by boiling is not commendable way as this will concentrates the nitrate ion in solution. The local government cooperating with local health sector should distribute information related to nitrate risk as periodically monitored by the local health center via the health sector should distribute information related to nitrate risk as periodically monitored by the local health center via the health center of community. Additionally, training and announcement should be broadly publicized, especially for uneducated people as well as for farmers. Nitrate risks can be mitigated through several following possible techniques [34].

- Sewage system management (i.e., be treated before discharged, install environment sound sewage facilities, respect effluent disposal guideline, train and educate local administration, formulate awareness campaigns for the general public)
- Environmental protection policies and regulations (i.e., governmental policy measures like Nitrate Directive, Water Framework Directive, Groundwater Directive, etc.)
- Drinking water standards (i.e., WHO and EPA guidelines)
- Prevention method (i.e., limit the amount of nitrogen applied, avoid over-irrigation, etc.)
- Beneficial management practices (BMP) for agriculture (i.e., use nitrogen fertilizer as efficiently as possible via better timing and placement, remove key sources of nitrogen)
- Prevention techniques (i.e., building manure storage in concrete pits, install a slurry-store)
- Water treatment techniques (i.e., blending drinking water, ion exchange, reverse osmosis, biological denitrification, in-situ treatment)

V. CONCLUSIONS

The study revealed that the contamination of nitrate in the study area is concerned with land-uses and anthropogenic acts. The source of nitrate contamination in shallow groundwater in the Kataan up to Negombo can be inferred to derive from on-site sanitations under denser population, which is followed by agriculture as secondary significance. The evidence from ratio of nitrate/chloride (1:1 – 8:1) also suggested that the primary nitrate contamination in water wells in the study area is of faecal origin due to improper on-site sanitations, while the rest may be from fertilizer applications. The estimated nitrate concentration in the area through application of equation suggested by Foster & Hirata [19] demonstrates a rough range of values as approximately less than 20 mg/L. Nonetheless, its usefulness is for predicting the probable nitrate content in groundwater under on-site sanitations. Recently, the analyzed groundwater samples indicated that 31% of the water samples are above the human affected value of 10 mg/L NO$_3^-$ though there are no samples exceeding 50 mg/L. Lower nitrate in the area may be due to denitrification processes. Although, the current condition of nitrate in the area is still found to be low, it will possibly increase if no proper management action to face with population growth rate and no on-site sanitations are adequately and properly established. Although this study has limited information, it may be considered as a good starting point and background value for identifying groundwater quality deterioration by nitrate.

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