ORIGINAL ARTICLE

Impact of pit-toilet leachate on groundwater chemistry and role of vadose zone in removal of nitrate and *E. coli* pollutants in Kolar District, Karnataka, India

Sudhakar M. Rao · M. Sekhar · P. Raghuveer Rao

Abstract Assessment of chemistry of groundwater infiltrated by pit-toilet leachate and contaminant removal by vadose zone form the focus of this study. The study area is Mulbagal Town in Karnataka State, India. Groundwater level measurements and estimation of unsaturated permeability indicated that the leachate recharged the groundwater inside the town at the rate of 1 m/day. The average nitrate concentration of groundwater inside the town (148 mg/L) was three times larger than the permissible limit (45 mg/L), while the average nitrate concentration of groundwater outside the town (30 mg/L) was below the permissible limit. The groundwater inside the town exhibited *E. coli* contamination, while groundwater outside the town was free of pathogen contamination. Infiltration of alkalis (Na^+, K^+) and strong acids (Cl^-, SO_4^{2-}) caused the mixed Ca–Mg–Cl type (60 %) and Na–Cl type (28 %) facies to predominate groundwater inside the town, while, Ca–HCO$_3$ (35 %), mixed Ca–Mg–Cl type (35 %) and mixed Ca–Na–HCO$_3$ type (28 %) facies predominated groundwater outside/periphery of town. Reductions in *E. coli* and nitrate concentrations with vadose zone thickness indicated its participation in contaminant removal. A 4-m thickness of unsaturated sand + soft, disintegrated weathered rock deposit facilitates the removal of 1 log of *E. coli* pathogen. The anoxic conditions prevailing in the deeper layers of the vadose zone (>19 m thickness) favor denitrification resulting in lower nitrate concentrations (28–96 mg/L) in deeper water tables (located at depths of −29 to −39 m).

Keywords *E. coli* · Groundwater · Nitrate · Pit toilets · Vadose zone

Introduction

Pollution of groundwater resources are geogenic and anthropogenic in origin. Contamination of groundwater by fluoride, arsenic and dissolved salts is mainly contributed by geological activities. Contamination of groundwater resources by organics, heavy metals, cyanides, aluminum and nitrates is anthropogenic in origin and arises due to uncontrolled discharges from industries, sewage treatment plants and agricultural applications of fertilizers and pesticides (Stamatis et al. 2011; Petrini et al. 2011; Rao et al. 2008). In addition, infiltration of pit-toilet leachates is an alarming source of anthropogenic contamination in India (Rao 2011).

The inorganic and microbial pollutants produced in pit toilets are removed by (1) physical and biochemical processes occurring at the biological mat or clogging zone at the interface of the pit and soil, (2) during leachate percolation through the vadose zone and 3) during storage and transport of leachate in the local aquifer (Gerba et al. 1991; Wilson et al. 1995; Howard et al. 2006; Leonard and Gilpin 2006; Miller et al. 2006; Parten 2010). Water travels slowly through the vadose zone; hence, on-site sanitation systems largely rely on this zone to treat the leachate contaminants by physico-chemical and bio-chemical processes. The ability of the vadose zone to remove contaminants is region specific. Pre-dominant presence of sands–gravel deposits in the vadose zone would facilitate quicker flow of leachate

S. M. Rao (✉) · M. Sekhar · P. Raghuveer Rao
Department of Civil Engineering,
Centre for Sustainable Technologies,
Indian Institute of Science,
Bangalore 560012, India
e-mail: msrao@civil.iisc.ernet.in

and lesser contaminant removal than the impermeable clay–silt deposit. Studies in different geographical regions are therefore necessary to enhance understanding on leachate–vadose zone–groundwater chemistry interactions.

This paper examines the impact of percolation of pit-toilet leachate on groundwater chemistry in residual soil deposit formation in Kolar District, Karnataka. The specific study area is located in Mulbagal Town, Kolar District. This town relies on pit toilets for disposal of human waste and on groundwater for its potable water requirements. The depth of groundwater table ranges from -8 to -39 m. As the bedrock above the water table is unsaturated, the groundwater depth also signifies the thickness of the vadose (unsaturated) zone at the particular location.

Study area

Mulbagal Town is located in Mulbagal Taluk, Kolar District, Karnataka State, India (Fig. 1). The town is located at a distance of 95 km from Bangalore. The town geographically lies between $78^{o}4'$ and $78^{o}24'$ E longitude and $13^{o}17'$ and $13^{o}10'$ N latitude and has an average elevation of 827 m (2,713 feet). Mulbagal Town has a geographical area of 8.5 km^2 and a population of about 60,000. The town experiences temperature variation between 18 and 35 °C (during winter and summer seasons) and receives an

average annual rainfall of 818 mm and rains on 72 days in a year.

Kolar District consists of immense expanses of migmatitic gneisses and younger granites that are seen as elongated NS trending patches intruding the gneisses. The gneissic complex is composed of composite gneisses, migmatites, granites and quartz veins. The weathered zone in the crystalline formation ranges from <1 to 20 m in thickness; larger thickness of weathered zones is encountered in valley portions over gneiss deposits (Report on Dynamic Groundwater Resources of Karnataka 2005). The fracture/fissure system developed along joints and faults traversing the rock facilitates groundwater circulation and holds moderate quantities of water (Jal Nirmal Project Report 2004). The transmissivity of the formation ranges from 2 to 1,935 m^2/day. The general yield of the formations varies from 0.8 to 30 l/s (Jal Nirmal Project Report 2004; DMG and CGWB 2005).

Materials and methods

Description of drinking water wells

Groundwater samples from 69 drinking water wells were examined from the study area. The majority of the wells were drilled between 2000 and 2005 to depths ranging
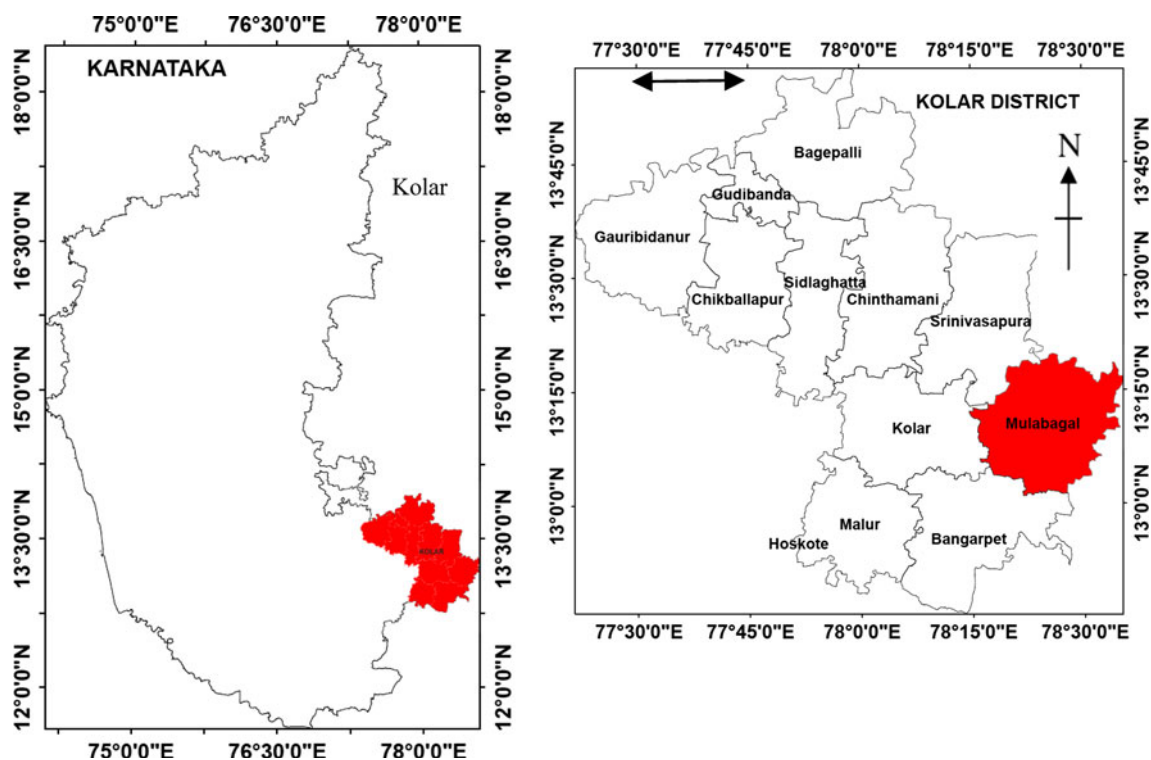

Fig. 1 Location of Mulbagal Town and distribution of drinking water wells

from 16 to 70 m. Based on the spatial distribution, the wells in Mulbagal Town are classified under two series. The inner town series (ITS) drinking water wells (43 number) are mostly located inside the town and pump-house series (PHS) wells (26 number) are located outside or the periphery of the town. The numerical designations of the PHS and ITS wells, their approximate depths (wherever data are available, data provided by the Town Municipal Council of Mulbagal) and static groundwater levels (measured on 20 April, 20 May and 1 June 2009 using Heron skinny dipper) are reported in Tables 1 and 2, respectively. The static state of the groundwater levels were ensured by allowing the water levels to recover for about 6–10 h after pumping from the concerned wells was stopped. This recuperation period was arrived at by monitoring groundwater levels of the wells in the post-pumping stage during preliminary field trials. The yields of the wells were measured to vary from 0.3 to 5.4 l/s. Drinking water wells belonging to the PH series supply water (5 million l/day) to centralized municipal supply system where pumped water is collected in sumps; water from the sumps are pumped to service reservoirs, which then supply water through piped network to individual households or community taps at street levels. In order to augment supply, drinking water wells were installed inside the town (belonging to the IT series); the water pumped from ITS wells directly feed into a separate localized pipe network that provides water to individual households/community taps/small water tanks (approximately 1,200 l capacity) in the vicinity of the well.

Sample collection

Collection of the groundwater samples from the 69 drinking water wells for laboratory testing was accomplished in five phases between April and June 2009 (Table 3). The intervals between sampling phases were mainly governed by availability of field personnel who assisted in water collection from the wells. During water collection, the junction between the well and the pipe leading to storage tanks were opened; groundwater was pumped out for about 15 min, following which samples were collected for laboratory analysis. Water samples for microbial examination collected in sterilized glass bottles were immediately stored in dry ice after collection, transported to laboratory on the day of collection and preserved at 4 °C. Water samples for chemical analysis were separately collected in 1-l capacity polyethylene bottles and were preserved at 4 °C after transportation to the laboratory. Before using the poly-ethylene bottles for sample collection, it was verified by laboratory experiments that the inner surface of polymer bottles are incapable of adsorbing inorganic ions. It may be noted that since Mulbagal Town is located at a distance of 100 km from Bangalore, each round of sample collection

and transportation to laboratory (located at Bangalore) required 12 h.

Laboratory analysis

All laboratory testing was initiated within 24 h of field collection. Laboratory analysis of water samples collected during each sampling round was accomplished in about 7 days. Water samples collected during each round of sampling were examined for total coliform (TC) and *E. coli* by multiple fermentation tube method (MPN—Most Probable Number technique) (IS 1622 1981; IS 5401 2002; APHA 1999). The pH and electrical conductivity (EC) of the collected water samples were measured in the field using a portable pH meter and electrical conductivity meter. The EC values are converted to total dissolved solids (TDS) using an approximate relation (Todd 1980):

$$1 \, (mg/L) = 1.56 \, (\mu Siemen/cm) \tag{1}$$

Equation 1 is valid for most natural waters having conductance in the range of 100–5,000 µSiemen/cm (Todd 1980). The concentrations of magnesium, calcium, sodium and potassium ions in the groundwater samples were determined using Thermo-ICAP 6500 inductively coupled plasma-optical emission spectrometer (ICP-OES). The minimum detection limits for calcium, magnesium, sodium and potassium ions for the Thermo-Cap 6500 ICP-OES corresponds to 0.003, 0.003, 0.06 and 0.13 mg/L respectively. Calibration standards of 1, 5 and 10 mg/L were used in the measurement of calcium, magnesium, sodium and potassium ions. Concentrations of sulfate, chloride and nitrate ions were determined using Dionex ICS 2000 ion chromatography system configured with hydroxyl based anion retention column. The minimum detection limits for fluoride, chloride, nitrate and sulfate ions using Dionex ICS 2000 ion chromatography system correspond to 0.0023, 0.0025, 008 and 0.006 mg/L, respectively. Calibration standards of 1, 5 and 10 mg/L were used in chloride measurements. Calibration standards of 1, 10 and 20 mg/L were used in nitrate and sulfate measurements. Calibration standards of 0.2, 1 and 2 mg/L were used in fluoride measurements. Bicarbonate ion concentration was determined using Metrohm 877 Titrino Plus Titrator. The minimum detection limit for bicarbonate measurement by Metrohm 877 Titrino Plus Automatic Titrator corresponds to 2 mg/L.

The total hardness (TH, mg/L $CaCO_3$) of the water samples was determined from the equation (Todd 1980):

$$TH = 2.5Ca^{2+} + 4.1Mg^{2+}. \tag{2}$$

The correctness of the chemical analysis data was performed by determining the cation/anion balance (CAB) for each well water sample. The CAB value was generally

Table 1 Well designation, groundwater elevation and depth of ITS wells

Well designation	Groundwater elevation (m/period)			Average groundwater depth (m)	Well depth (m)	Dominant rock formation[a]
	20 April 2009	20 May 2009	1 June 2009			
MBW114	−29.1	−45.0	−35.1	−36.4	NA	Gneisses and unclassified crystallines
MBW117	−15.0	−15.5	−14.0	−14.8	−32	Gneisses and unclassified crystallines
MBW120	−9.0	−11.2	−10.0	−10.1	−66	Gneisses and unclassified crystallines
MBW115	−32.0	−43.1	−37.0	−37.4	NA	Gneisses and unclassified crystallines
MBW116	−17.1	−18.0	−19.0	−18.0	−37	Gneisses and unclassified crystallines
MBW118	−12.0	−15.4	−16.3	−14.6	−17	Gneisses and unclassified crystallines
MBW119	−17.1	−19.5	−18.0	−18.2	−32	Gneisses and unclassified crystallines
MBW121	−21.7	-38.9	-35.0	-31.9	NA	Gneisses and unclassified crystallines
MBW212	−32.9	−35.1	−30.0	−32.7	NA	Gneisses and unclassified crystallines
MBW224	−32.0	−33.2	−30.0	−31.7	−35	Gneisses and unclassified crystallines
MBW225	−8.5	−10.0	−8.6	−9.0	−23	Gneisses and unclassified crystallines
MBW227	−11.5	−12.1	−12.5	−12.0	−27	Gneisses and unclassified crystallines
MBW228	−7.8	NA	−10.0	−8.9	−37	Gneisses and unclassified crystallines
MBW229	1−0.0	−9.8	−10.0	−9.9	−16	Gneisses and unclassified crystallines
MBW230	−7.3	−8.0	−7.0	−7.4	−39	Gneisses and unclassified crystallines
MBW231	−7.4	−8.0	−8.2	−7.9	−19	Gneisses and unclassified crystallines
MBW233	−13.5	−13.7	−13.6	−13.5	−41	Gneisses and unclassified crystallines
MBW234	−9.0	−11.0	−7.0	−9.0	−17	Gneisses and unclassified crystallines
MBW235	−31.0	−31.5	−28.5	−30.3	−37	Gneisses and unclassified crystallines
MBW236	−30.0	−32.2	−24.8	−29.0	−55	Gneisses and unclassified crystallines
MB239	−31.6	−33	−40.1	−34.9	NA	Gneisses and unclassified crystallines
MB240	−32.0	−32.6	−39	−34.5	−89	Gneisses and unclassified crystallines
MBW244	−40.0	−34.0	−43.0	−39.0	NA	Gneisses and unclassified crystallines
MBW245	−4.0	−4.7	−6.5	−5.1	−46	Gneisses and unclassified crystallines
MBW300	−32.5	−33.4	−33.5	−33.1	NA	Gneisses and unclassified crystallines
MBW312	−6.8	−6.0	−4.5	−5.8	−23	Gneisses and unclassified crystallines
MBW313	−15.8	−17.4	−13.4	−15.5	−37	Gneisses and unclassified crystallines
MB314	NA	NA	NA	NA	NA	Gneisses and unclassified crystallines
MBW400	−13.2	−14.0	−14.5	−13.9	−17	Gneisses and unclassified crystallines
MBW401	−13.0	−13.6	−15.0	−13.9	−41	Gneisses and unclassified crystallines
MBW402	−9.9	−11.2	−15.5	−12.2	−37	Gneisses and unclassified crystallines
MBW403	−10.5	−11.7	−12.0	−11.4	NA	Gneisses and unclassified crystallines
MBW404	−18	−19.0	−20	−19.0	−29	Gneisses and unclassified crystallines
MBW405	−11.5	−13.6	−12.0	−12.4	NA	Gneisses and unclassified crystallines

Table 1 continued

Well designation	Groundwater elevation (m/period)			Average groundwater depth (m)	Well depth (m)	Dominant rock formation[a]
	20 April 2009	20 May 2009	1 June 2009			
MBW406	−12.0	NA	−12.5	−12.3	NA	Gneisses and unclassified crystallines
MB407	NA	NA	NA	NA	NA	Gneisses and unclassified crystallines
MBW408	−8.0	−8.9	−10.0	−9.0	−37	Gneisses and unclassified crystallines
MBW410	−10.2	−10.5	−11.0	−10.6	−37	Gneisses and unclassified crystallines
MBW411	−11.0	−12.2	−11.0	−11.4	−32	Gneisses and unclassified crystallines
MB413	NA	NA	NA	NA	NA	Gneisses and unclassified crystallines
MBW415	−9.6	−10.0	−8.5	−9.4	−37	Gneisses and unclassified crystallines
MBW419	−8.0	−8.7	−10.0	−8.9	NA	Gneisses and unclassified crystallines
MBW421	−14.5	−14.3	−15.0	−14.6	−44	Gneisses and unclassified crystallines

NA not available

[a] DMG and CGWB (2005)

less than ± 5 % for the total sample set and did not exceed ± 10 % for any one sample. The cation/anion balance (CAB, %) is calculated from the equation:

$$\text{CAB } (\%) = \frac{(\text{sum of cations}) - (\text{sum of anions})}{\text{sum of cations} + \text{sum of anions}} \times 100. \tag{3}$$

The probability distribution function (PDF) for a specified concentration range (x) of a given contaminant is obtained from the equation:

$$\text{PDF } = f(x, \mu, \sigma) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\left(\frac{[x-\mu]^2}{2\sigma^2}\right)} \tag{4}$$

where μ is the average and σ is the standard deviation of all concentration values of a given contaminant belonging to a well series (ITS or PHS). PDF is computed for a range of x values for a given contaminant. A plot of PDF versus concentration range (x) of the given contaminant is generated. The plot is interpreted to predict the probability of concentration of the given contaminant exceeding the desirable/permissible limit for drinking water in a particular well series. The contaminants examined are TDS, nitrates, *E. coli* and TH.

Results and discussion

E. coli, nitrate and salt contamination

Table 4 summarizes the minimum to maximum range, average concentration of contaminant, standard deviation from mean and percentage of samples exceeding the desirable and permissible limits (IS 10500 2003) for nitrate, *E. coli*, TDS and TH (total hardness expressed as $CaCO_3$) contaminants in the drinking water wells. The nitrate concentrations in ITS wells range from 4 to 388 mg/L with a mean value of 148 mg/L. Further, 79 % of water samples exhibit nitrate concentrations in excess of 45 mg/L (permissible limit in drinking water = 45 mg/L, IS 10500 2003). Comparatively, nitrate concentrations in PHS wells range from 1 to 115 mg/L with a mean value of 30 mg/L and only 13 % of well samples exhibit nitrate concentrations >45 mg/L. The *E. coli* levels in the ITS wells range between 0 and 1,601 MPN/100 mL (MPN = most probable number) with mean value of 189 and 55 % of the samples exhibiting pathogen contamination. The PHS wells show much lesser *E. coli* contamination with values ranging between 0 and 6 MPN/100 mL, an average value of 1 MPN/100 mL, and only 9 % of samples exhibit *E. coli* contamination. The ITS wells are characterized by TDS concentrations of 254–1,883 mg/L with an average value of 1,057 mg/L, and 83 % of samples exhibit dissolved salts concentration above the desirable limit for drinking water

Table 2 Well designation, groundwater elevation and depth of PHS wells

Well designation	Groundwater elevation (m/period)			Average groundwater depth (m)	Well depth (m)
	20 April 2009	20 May 2009	1 June 2009		
MB101	−27.1	−27	−28.2	−27.4	NA
MB107	−19.1	NA	−21.3	−20.2	NA
MB108	−30.0	−30.5	−33	−31.2	NA
MB110	−23.6	−27	−27.1	−25.9	−69
MBW122	−6.5	−33.0	−10.0	−16.5	NA
MBW123	−37.0	−37.1	−41.0	−38.4	NA
MBW124	−39.1	−40.0	−48.2	−42.4	NA
MB200	−12.0	−12.9	−13.5	−12.8	NA
MB202	−13.0	−17.5	−20	−16.8	−67
MB203	−25.0	−24.5	−24.5	−24.7	NA
MB205	−13.3	−15	−16	−14.8	NA
MB206	−16.7	−18.5	−14	−16.4	−32
MB207	−16.5	−18.2	−17.5	−17.3	NA
MB209	−33.6	−36.2	−35.8	−35.2	NA
MB210	−33.0	NA	−33.5	−33.3	NA
MB217	−21	−23.2	−25	23.1	−70
MB221	−29.0	−30	NA	−29.5	−39
MB237	−			−	NA
MB304	−18.0	−18.5	−17.9	−18.1	−49
MB305	−19.5	−19.8	−19.8	−19.7	−51
MB306	−24.6	−25.6	−25	−25.1	−62
MB308	−25.0	−26.1	−28	−26.4	−73
MB309	−11.5	−13	−11	−11.8	−17
MB310	−12.1	−13.6	−15	−13.6	−48
MB316	−36.5	−42.2	−37.5	−38.7	−48
MB417	−7.5	−8.0	−9.0	−8.2	NA

Table 3 Collection schedule of groundwater samples

Collection date	Number of groundwater samples collected
15 April 2009	20
28 April 2009	10
13 May 2009	19
3 June 2009	11
25 June 2009	9

(500 mg/L, IS 10500 2003). The drinking water wells located outside the town (PHS wells) are less saline (TDS ranges from 321 to 1,280 mg/L) and exhibit lower average TDS value (679 mg/L); 61 % of wells have TDS concentrations in excess of the desirable limit. The wells located outside the town are less saline as they are not exposed to leachate infiltration from pit toilets. The total hardness (TH, expressed as $CaCO_3$) of drinking water wells located inside the town (IT series) ranges from 78 to 858 mg/L with an average value of 525 mg/L. Forty-two percent of ITS wells exhibit TH values in excess of the desirable limit

(300 mg/L) and 39 % wells exhibit TH values in excess of the permissible limit. The TH values of the PHS wells range between 187 and 892 mg/L with an average value of 395 mg/L; 59 % of wells exhibit TH in excess of the desirable limit and 9 % exhibit TH values in excess of the permissible limit. The data in Table 4 also reveal that contaminant concentrations show larger scatter from the mean (larger standard deviation) for wells located inside the town (ITS wells) than for those located outside/periphery of town (PHS wells). The water quality data are analyzed using the probability density function (PDF) technique (for normal distribution) to evaluate the likelihood of well contamination.

Figures 2, 3 and 4 present the probability density function for *E. coli*, nitrate and TDS contamination in drinking water wells belonging to the IT and PH series. The PDF plots in Fig. 2 indicate that there exists 100 % probability that drinking water wells located outside the town (PH series) are free of pathogen contamination. Comparatively, there exists only 32 % probability that drinking water wells located inside the town (ITS Series) would not

Table 4 Statistical summary of water quality parameters

Parameter	Minimum	Maximum	Mean	Standard deviation (SD)	% Samples exceeding permissible limit	% Samples exceeding desirable limit
Nitrate (ITS) (mg/L)	4	388	148	99	79 (45)	Not applicable
Nitrate (PHS) (mg/L)	1	115	30	27	13	Not applicable
E. coli (ITS) (MPN/100 mL)	0	1,601	189	409	55 (0)	Not applicable
E. coli (PHS) (MPN/100 mL)	0	6	1	1.48	9	Not applicable
TDS (ITS) (mg/L)	254	1,883	1,057	443	Nil (2000)	83 (500)
TDS (PHS) (mg/L)	321	1,280	679	285	Nil	61 (500)
TH (ITS) (mg/L	74	858	525	546	42 (300)	36 (600)
TH (PHS) (mg/L)	187	892	395	177	59 (300)	9 (600)

Values in parenthesis represent permissible or desirable limits as per IS 10500 (2003)
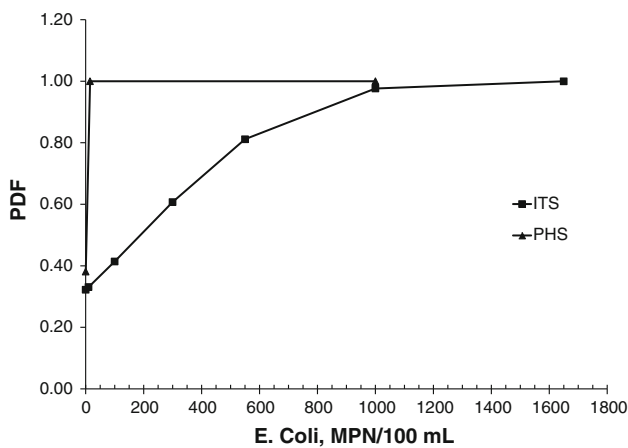
Fig. 2 Probability density function of *E. coli* contamination in drinking water wells belonging to the IT and PH series

Fig. 4 Probability density function of dissolved salt (TDS) contamination in drinking water wells belonging to the IT and PH series

Fig. 3 Probability density function of nitrate contamination in drinking water wells belonging to the IT and PH series

exhibit *E. coli* contamination (Fig. 3). The PDF plots in Fig. 3 illustrate that there exists 71 % probability that the PHS wells would not exhibit nitrate contamination

($NO_3^- < 45$ mg/L); however the probability of nitrate contamination being absent in ITS wells is mere 14 %. The PDF plots in Fig. 4 illustrate that there exists 28 % probability that total dissolved solids (TDS) concentration in PHS wells will be <500 mg/L (desirable limit for drinking water); comparatively, the probability of TDS concentration in ITS wells being <500 mg/L is only 7 %. The much larger probability of *E. coli* and nitrate contamination of the drinking water wells located inside Mulbagal Town (IT series) is a consequence of leachate infiltration from pit toilets to groundwater as will be elucidated in the next sections.

Hydrogeochemical facies and mechanisms controlling groundwater chemistry

Figures 5 and 6 present the Piper plots for groundwater samples from PHS and ITS wells. The distribution of data points in the lower base triangles reveals that the majority of the samples from PHS and ITS wells cannot be
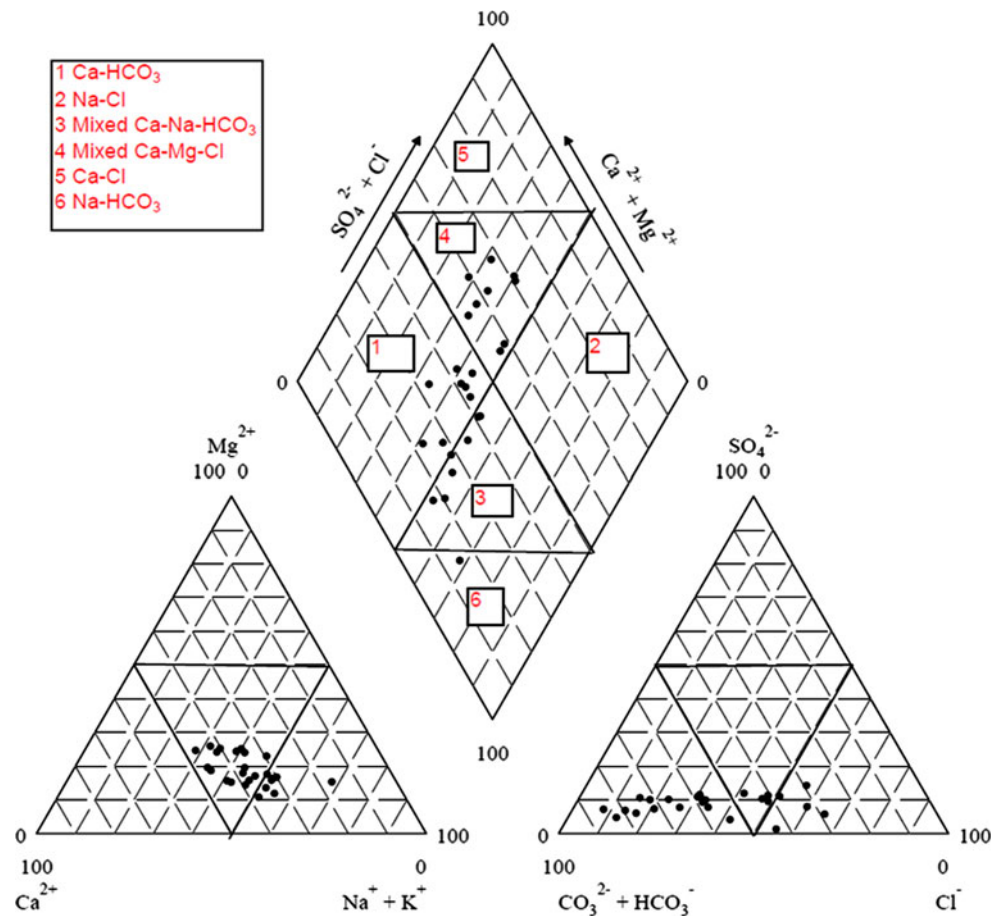
categorized into any dominant cation type. A majority of
PHS samples (around 65 %) categorize as HCO_3 type,
16 % as Cl type and the remaining samples cannot be
classified into any particular category. Comparatively,
44 % of ITS samples may be categorized as Cl type, 14 %
as HCO_3 type and the remaining cannot be classified into
any particular category. The distribution of data points in
rhomboids in Fig. 5 reveals that equal fractions of PHS
groundwater samples fall in the fields of Ca–HCO_3 (35 %)
and mixed Ca–Mg–Cl (35 %) types and about 28 % of
samples are mixed Ca–Na–HCO_3 type. The distribution of
data points in lower base triangles in Fig. 5 reveals that the
PHS samples cannot be categorized into any dominant
cation type, and weak acids (HCO_3^- and CO_3^{2-}) dominate
over strong acids (Cl^- and SO_4^{2-}). Comparatively, 60 %
of ITS groundwater samples may be categorized as
Ca–Mg–Cl type, 28 % as Na–Cl type and the remaining are
distributed among mixed Ca–Na–HCO_3 and Ca–Cl types
(Fig. 6). The plot indicates enrichment of groundwater
inside the town by alkalis (Na^+ and K^+) and strong acids
(Cl^- and SO_4^{2-}) from leachate infiltration.

The source of dissolved ions in groundwater samples can
be broadly assessed from the Gibb's diagram by plotting the
ratio of Na/(Na + Ca) and Cl/(Cl + HCO_3) as a function of

total dissolved solids (TDS) (Gibbs 1970). Based on the
zones where the cationic (Fig. 7a) and anionic (Fig. 7b)
ratios plot, the processes controlling the chemistry of
groundwater samples is classified as: (1) evaporation–crys-
tallization dominance (TDS > 800 mg/L), (2) rock-weath-
ering dominance (TDS, 40–800 mg/L) and (3) atmospheric
precipitation dominance (TDS < 40 mg/L). The distribu-
tion of data points in Fig. 7a, b suggests that rock-weathering
mechanism controls the chemistry of groundwater outside
the town (PHS samples), while, evaporation–crystallization
controls the chemistry of groundwater inside the town (ITS
samples). Kolar District is characterized by semi-arid cli-
mate, and soils in these regions are formed by in situ
weathering of parent rock (Rao and Venkatesh 2012). As
water is the principal agent for chemical weathering of rocks,
rock weathering controls the groundwater chemistry in Kolar
District. The Gibbs diagram accordingly shows that rock
weathering controls the chemistry of groundwater samples
outside the town. In the absence of leachate contamination,
rock weathering should also have controlled the chemistry of
groundwater inside the town. Enrichment of groundwater by
alkalis (Na^+ and K^+) and strong acids (Cl^- and SO_4^{2-}) from
leachate infiltration cause data points of ITS wells to plot in
the evaporation–crystallization zone.

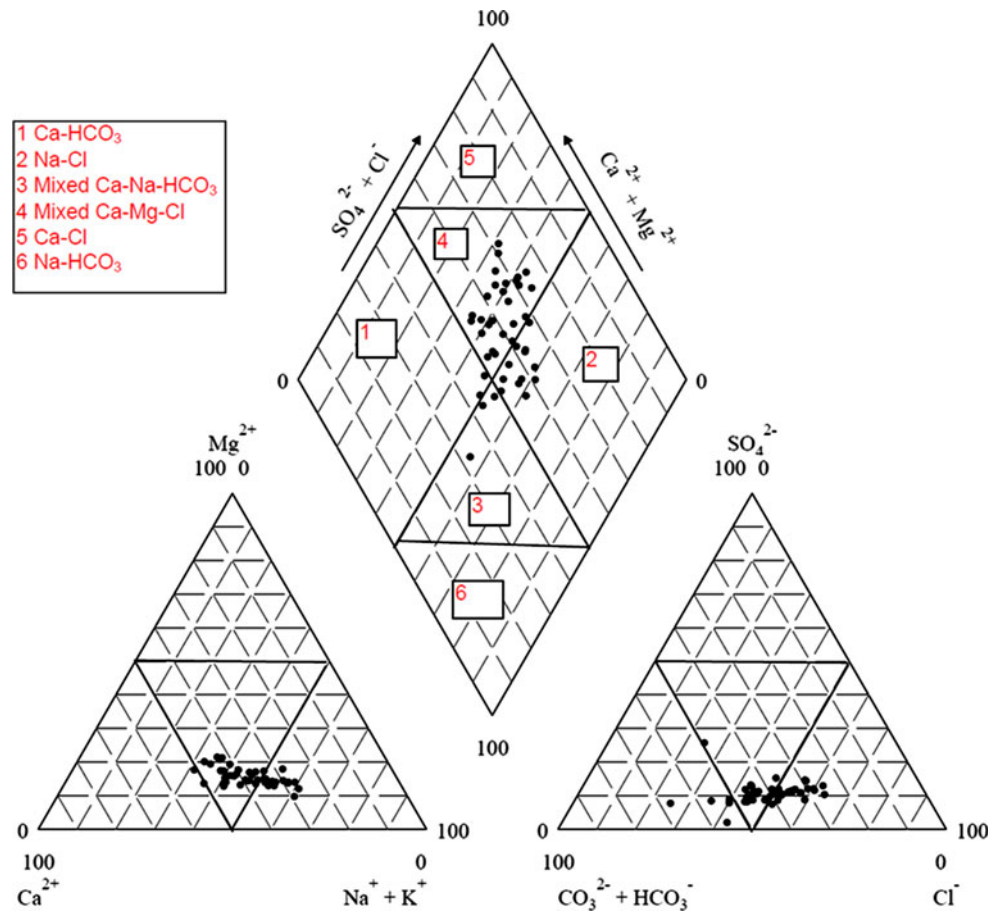**Fig. 6** Piper plot for groundwater samples from ITS wells

Figure 8 presents the frequency histograms for groundwater depth in the ITS and PHS wells. The histograms in Fig. 8 indicate that drinking water wells located inside the town are subject to recharge than evaporation, as the frequency of wells (31 of 43) having lower range of groundwater depth (<-20 m) outnumber the frequency of wells (4 of 43) with maximum depth (>-36 m). Recharge of groundwater by the pit-toilet leachate than evaporation–crystallization hence contributes to groundwater chemistry inside the town.

Contaminant removal by the vadose zone

Figure 9 plots log10 removal of *E. coli* as a function of the vadose zone thickness for drinking water wells located inside the town. The data for drinking water wells located outside the town are not considered as they are free of pathogen contamination. As previously stated, the average groundwater depth of a given well signifies the thickness of the vadose zone at a particular location. The occurrence of partially saturated soil voids causes water to flow relatively slowly in the vadose zone (Lu and Likos 2004). The slow velocity restricts the distance over which the bacteria travel in the vadose zone by facilitating their removal through

biological filtration, adsorption on surface of soil particles and death (Cave and Kolsky 1999).

The *E. coli* concentration in raw sewage is about 1,000,000 MPN/100 mL; consequently, the drinking water standard of zero *E. coli*/100 mL necessitates seven log removal of the pathogen (Leonard and Gilpin 2006).

The *x*-axis values in Fig. 9 are calculated as:

$$\log_{10} 1,000,000 \frac{\text{MPN}}{100\,\text{mL}} - \log_{10} \times \frac{\text{MPN}}{100\,\text{mL}} \tag{5}$$

where *x* represents the *E. coli* value in the drinking water well belonging to the IT series. The regression equation in Fig. 9 illustrates that on an average, 4-m thickness of vadose (unsaturated) zone facilitates 1 log removal of *E. coli* pathogen. Consequently locations with water table depths ≥ -25 m are free from *E. coli* contamination in Fig. 9. Studies are available which suggest that about 300–900 mm of many soil types provides high levels of pathogen reduction (Leonard and Gilpin 2006; Parten 2010). According to Stevika et al. (2004), the presence of organic and clay matter in a porous media renders it favorable for pathogen removal by filtration, adsorption and adhesion to biofilms. Borehole profiles in residual soil deposits that are typical of Mulbagal Town exhibit few
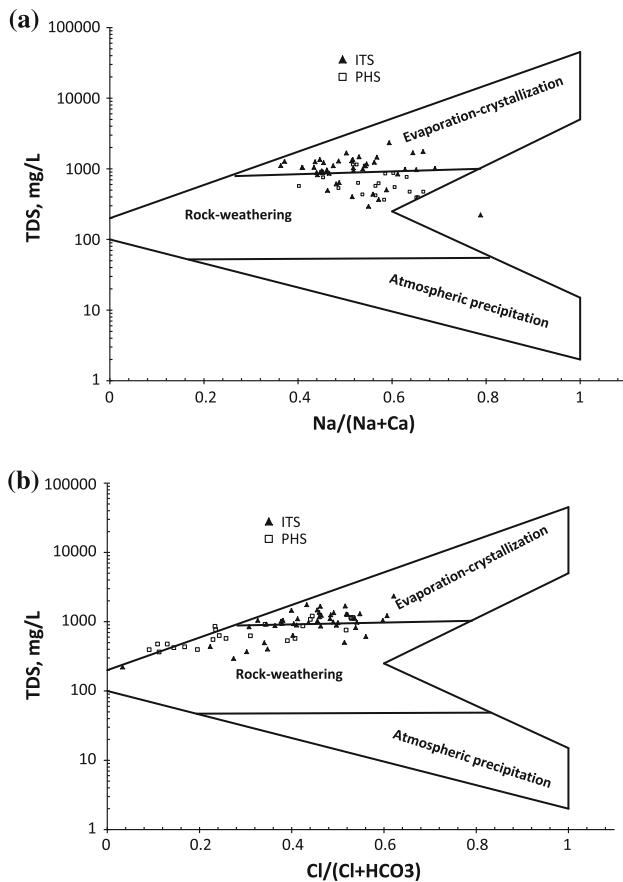
(a)

(b)

Fig. 7 a, b Gibbs diagram for groundwater samples from PHS and ITS wells
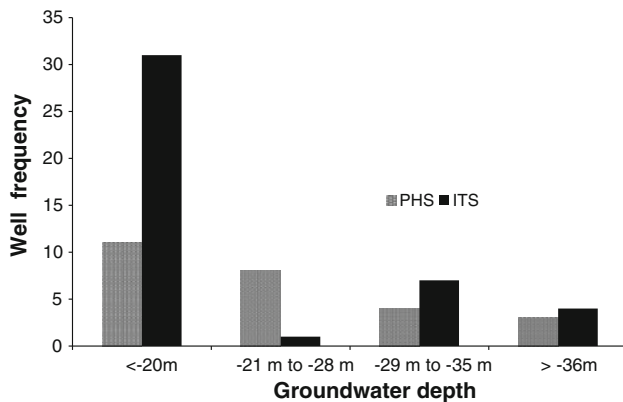

Fig. 8 Frequency distribution of groundwater depth in PHS and ITS wells

meters (0–6 m) of reddish-brown sandy clay–silt (silt + clay = 55–65 %, sand = 35–45 %), followed by sandy deposits (sand = 60–75 %, gravel = 0–18 %, silt + clay = 25–40 %) of variable thickness that in turn is underlain by soft, disintegrated weathered rock (Rao and Venkatesh 2012). The large thickness (4 m) of the vadose zone needed for 1 log removal of pathogen in the present
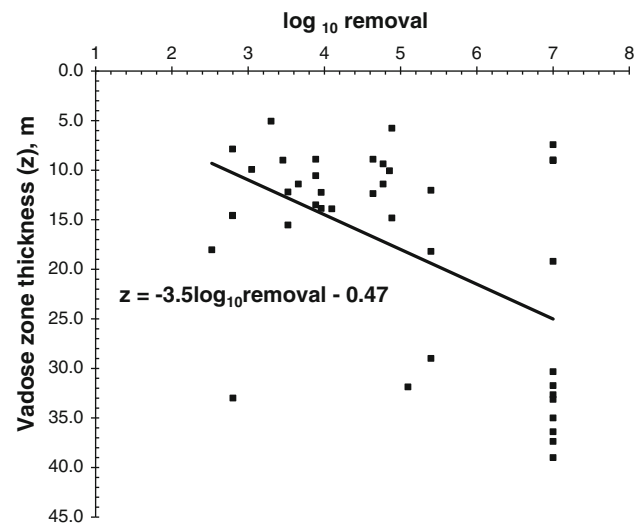

Fig. 9 Variation of *E. coli* concentration with vadose zone thickness

study suggests the paucity of organic matter and clay content in the unsaturated sand and soft-disintegrated rock deposit. In a related study in India, at a site characterized by sandy soil deposit with groundwater velocity of 0.9 m/day, a horizontal separation of 7.5 m between borehole latrine and open well was recommended as an ample margin of safety against bacterial contamination (Lewis et al. 1980).

Figure 10 plots nitrate concentration in ITS well samples as a function of the vadose zone thickness. Again, the average water table depth (Table 1) signifies the vadose zone thickness at a particular location. It is observed that vadose zone thickness of up to 19 m does not produce any trend in nitrate concentrations in the groundwater. The nitrate concentrations vary between 5 and 390 mg/L with the majority of values clustered between 120 and 290 mg/L. The large differences in nitrate concentrations at shallower groundwater depths (≤ -19 m) are attributed to variability in nitrate loadings from the pit-toilet leachates.

Vadose zone depths larger than 29 m apparently produce lesser groundwater nitrate concentrations with values ranging from 28 to 96 mg/L (Fig. 10). The maximum vadose zone depth in Mulbagal Town corresponds to 39 m. The travel rate of nitrate contaminated leachate in the vadose zone can be estimated from the hydraulic conductivity of this deposit. The hydraulic conductivity of unsaturated soil deposits (k_{unsat}) can be estimated using an empirical equation (Fredlund et al. 1994):

$$k_{unsat} = k_s \left(\frac{\theta}{\theta_s} \right)^n \tag{6}$$

where k_s is the permeability of saturated material, θ is the volumetric water content, θ_s is the volumetric water content at saturation and n is shape factor. The values of k_s, θ_s and n for sand (material comprising the vadose zone)
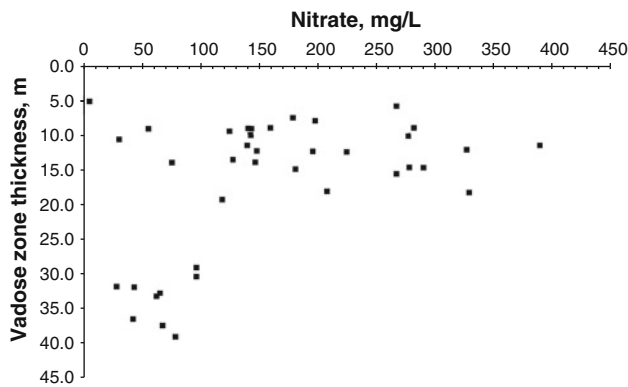
Fig. 10 Variation of nitrate concentration with vadose zone thickness

correspond to 642.9 cm/day, 0.37 and 3.18, respectively (Carsel and Parrish 1988). The θ value for sand deposit in the vadose zone is calculated as:

$$\theta = \frac{\theta_s + \theta_r}{2} \tag{7}$$

where θ_r is residual water content (0.05 for sand) and θ_s is the saturated water content (0.37 for sand, data from Carsel and Parrish 1988). Inserting appropriate values of θ (0.21), k_s, θ_s and n in Eq. 6 yield unsaturated permeability (k_{unsat}) of 1 m/day for the vadose zone. The k_{unsat} value implies that the pit-toilet leachate contaminated with *E. coli* and dissolved salts travels at a rate of 1 m/day in the vadose zone. Consequently after 39 days of travel, the groundwater located at a depth of −39 m is susceptible to nitrate contamination from leachate infiltration.

Despite the relatively short period (39 days) required by the leachate to contaminate the deepest groundwater table (located at −39 m depth), the lower nitrate concentrations (28–96 mg/L) in deeper water tables (located at depths of −29 to −39 m) than shallower ($\leq$ −19 m) water tables (120 and 290 mg/L) suggest that denitrification is favored in the deeper vadose zone. The extent of denitrification process in the vadose zone is linked to the absence or near absence of oxygen, an adequate supply of electron donor (carbon) and capable bacterial population (Korum 1992; Miller et al. 2006). According to Brockman et al. (1992), besides transporting carbon, water also transports microbes below the surface to high recharge sites that harbor different culturable bacteria. It is probable that anoxic conditions conducive for denitrification are encountered at vadose zone depths >18 m, which in combination with carbon source and capable bacteria population cause reduction of nitrate according to the reaction Miller et al. (2006):

$$1/4\,CH_2O + 1/5NO_3^- + 1/5H^+$$
$$\rightarrow 1/10N_2\,(gas) + 7/20H_2O + 1/4CO_2\,(gas) \tag{8}$$
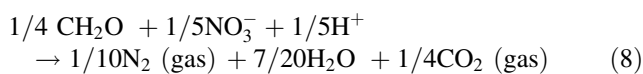
In case of inadequacy of carbon source, residual ammonia could act as reducing source for nitrates (Miller et al. 2006):

$$5/4NH_4^+ + 3/4NO_3^- \rightarrow N_2(gas) + 9/4H_2O + 1/2H^+ \tag{9}$$
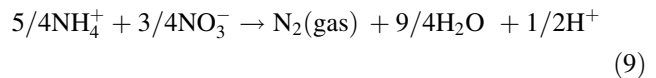
Measurements of stable isotopes and dissolved gases are used to support the denitrification process (Bohlke 2002); however, budgetary and field limitations prevented such a study.

Conclusions

Leachate infiltration from pit toilets has contaminated the drinking water wells located inside Mulbagal Town. The nitrate concentrations in ITS wells ranged from 4 to 388 mg/L with a mean value of 148 mg/L. The *E. coli* levels in the ITS wells ranged between 0 and 1601 MPN/100 mL with a mean value of 189 MPN/100 mL. Comparatively, the drinking water wells that were located outside the town were predominantly free of nitrate and pathogen contamination. The average TDS concentrations of ITS and PHS wells correspond to 1,057 and 679 mg/L; the PHS wells are less saline as they are free from leachate infiltration. The average TH values of ITS and PHS wells correspond to 525 and 395 mg/L indicating that leachate infiltration imposes additional calcium carbonate hardness to groundwater inside the town. Evaluation of the hydrogeochemical facies revealed that 35 % of PHS groundwater samples may be categorized as Ca–HCO$_3$, 35 % as mixed Ca–Mg–Cl type and about 28 % as mixed Ca–Na–HCO$_3$ type. Comparatively, infiltration of alkalis and strong acids caused the groundwater inside the town to be categorized as mixed Ca–Mg–Cl type (60 %) and Na–Cl type (28 %), and the remaining being distributed among mixed Ca–Na–HCO$_3$ and Ca–Cl types. As water is the principal chemical weathering agent, rock weathering controls the groundwater chemistry outside Mulbagal Town. In the absence of leachate contamination, rock weathering ought to have controlled the groundwater chemistry inside the town. Enrichment of groundwater by alkalis and strong acids causes data points of ITS wells to plot in the evaporation–crystallization zone. The frequency histograms of groundwater depths in ITS wells suggest that recharge of groundwater by pit-toilet leachate than evaporation–crystallization controls groundwater chemistry inside the town.

The relations between contaminant concentration and vadose zone thickness for drinking water wells located inside the town indicate that the vadose zone facilitates removal of *E. coli* and nitrate ions. It was observed that 4-m thick of unsaturated sand and soft-disintegrated rock

deposit remove 1 log of *E. coli* pathogen. Consequently, drinking water wells having water table depths ≥ -25 m were free from *E. coli* contamination. Calculations show that the pit-toilet leachate flows at a rate of 1 m/day in the vadose zone and requires 39 days to contaminate the groundwater located at a depth of -39 m. The distribution of nitrate concentrations (28–96 mg/L) in deeper (located at depths of -29 to -39 m) and shallower (≤ -19 m) water tables (120 and 290 mg/L) were non-uniform suggesting that anoxic conditions prevailing in the deeper layers of vadose zone favored denitrification.

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