

Fate of phosphate and nitrate in waters of an intensive agricultural area in the dry zone of Sri Lanka

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Abstract The chemistry of surface waters and groundwater draining agricultural catchments in the north-central and northwestern areas of Sri Lanka is described. Hydrochemical data from 296 water samples are used to evaluate water quality and to identify the processes that control nitrate and phosphate concentrations in the water. The results indicate that nutrient concentrations in the groundwaters are greater than those in the surface waters. Increased nutrient levels were observed in groundwater in a selected area in the fortnight following fertilizer application. Detailed geochemical investigations of selected groundwater samples reveal a gradual rise of nitrate–N and other solutes along the horizontal flow direction. Compared to the application rates of fertilizer in the area, the average nutrient concentrations in all waters are relatively low (1.5 mg/l nitrate and 0.5 mg/l phosphate) and stable. The results suggest that prevailing reducing conditions, iron-rich overburden soil cover and manmade canal networks control nutrient accumulation in the groundwater.

Keywords Groundwater and surface water · Nitrate · Phosphate · Fertilizer · Accumulation · Canal · Sri Lanka

Introduction

The use of fertilizer has changed the world dramatically in recent decades, mainly through water pollution by nutrients

originating from agricultural activity (Addiscott et al. 1991; Vidal et al. 2000). Intensive use of fertilizer for crops is responsible for nitrate and phosphate accumulation in both groundwater and surface waters (Strebel et al. 1989; Clenaghan 2003). Water bodies polluted by nutrients create many environmental problems (Spalding and Exner 1993), including algal blooms and eutrophication in aquifers or in surface water bodies, and may produce potential hazards to human health (Fan and Steinberg 1996; Gelberg et al. 1999; Gulis et al. 2002).

Sri Lanka has been an agriculturally based country for centuries, and hence inevitably faces the threat of groundwater contamination due to excess fertilization (Dissanayake 1988; Gunatilake and Gunatilake 2004; Silva De and Ayomi 2004). Irresponsible fertilizer use and excess application introduce large amounts of nutrients to water bodies, to the detriment of the environment (Dissanayake and Weerasooriya 1987). This is especially the case in paddy cultivation.

The north-central part of Sri Lanka has been heavily used for paddy cultivation, especially in the last few centuries. Forty percent of the north-central district has been used for paddy cultivation. In addition to this, banana, papaya, and vegetable cultivation also occurs. Information from government organizations and individual farmers shows that the fertilizer application is high, and may be six to ten times in excess of levels recommended by government. The quantity of urea fertilizer applied to paddies varies from 100 to 150 kg per acre, and triple super phosphate (TSP) applications range from 75 to 100 kg per acre. In banana and papaya plantations, 100 g of urea/TSP fertilizer per plant is added once every 3 months. Since the fertilizer application within the study area is remarkably high compared to recommended rates, the effect of fertilizer application on water quality has been considered in detail.

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Much of the local population depends on groundwater for drinking and domestic purposes (Dissanayake and Weerasooriya 1985). Groundwater potential in these zones is limited due to the limited storage and low permeability of the underlying crystalline rock formations. This study examines the nutrient contamination of groundwater and surface water in the highly agricultural north-central part of Sri Lanka. Furthermore, we focus on identifying the causative factors that control the chemical quality of the water.

Physiography, climate, and hydrology of the study area

The study area is mainly composed of flat lands with elevations of 100–400 m above sea level, forming an undulating peneplain. The highest elevations occur toward the south and the west. The topography is undulating, with ridges and hills that rise above the base elevation, and narrow alluvial flood plains along the rivers (Second interim report on Kala Oya Basin comprehensive plan 2002). The study area falls within the dry zone of Sri Lanka, which is noted for its very low rainfall (1,200–1,700 mm/annually) and high evapotranspiration (1,706 mm/annually) for a prolonged period. Most rainfall is confined to a short period from October to January, which accounts for more than 70% of annual precipitation.

The depth of the groundwater table varies from 1 to 10 m, with an average seasonal fluctuation of 4 m. The main groundwater-bearing formations in the area are fractured crystalline bedrocks and weathered overburden (Second interim report on Kala Oya Basin comprehensive plan 2002). Thickness of the overburden ranges from 1 to 20 m, with average thickness of about 12 m. Montmorillonite and kaolinite are the most abundant clay minerals in the soil cover (Herath 1998). Alluvial deposits along active streams also play a vital role in water supply.

Geological setting of the study area

The geology of the study area is dominated by high-grade Precambrian metamorphic rocks (Cooray 1995), with minor occurrences of carbonatite intrusions in the central part. The main metamorphic rocks are charnokitic gneiss, calc-gneiss, marble and quartzite. All these lithotypes show a general NNE-SSW strike direction, and dip moderately toward the west.

Canal network

The regolith aquifer coincides with areas where small tank cascade systems have existed since ancient times. These aquifers are closely linked with the surface water in

streams, canals, and tanks. Being shallow, the regolith aquifers are highly susceptible to agricultural and other forms of contamination.

Large networks of canals are used for irrigation purposes. These canals distribute water to almost all the paddy fields in the area. Since ancient times it has been the practice to use lakes that are filled by rainwater for irrigation and water. In the last four decades water has been fed to the lakes by irrigated canals or directly from precipitation, collected in a cascade of lakes. Almost 600 man-made lakes occur in the study area, and most are used for irrigation purposes.

Canals situated at the far ends of the paddy fields are not lined or plastered, and hence seepage of dissolved ions is high. At the distribution end, the canals are wide and well plastered, and flow rates are high; these canals become progressively narrower and flow rates fall. The lower valley area receives diversions from the Mahaweli basin, with unlined canals running on either side.

Methodology

Surface water and groundwater samples were collected from 289 locations for this study, with sites selected based on available data and initial field investigations (Fig. 1a). Surface water samples were collected from lakes, canals, and streams. Groundwater samples were collected from wells dug for domestic use (hereafter termed dug wells), from wells used for agricultural use (agricultural wells), and from deep groundwater wells (tube wells). The study was extended to monitor nutrient variations in groundwater with time, and from areas with different vegetation cover (Fig. 1b). Sampling was carried out at 20 sites on a weekly basis, although no samples were collected on heavy rainfall days in order to avoid dilution effects.

Conductivity, temperature, and pH of the water samples were measured during sample collection. Nitrate and phosphate contents were determined using a HACH DR 2010 spectrophotometer, and cations (Na, Ca, Fe, Mn, K, and Mg) were measured with a Perkin Elmer atomic absorption spectrophotometer at the Department of Geology, University of Peradeniya, Sri Lanka. Standard methods of sampling and analysis were used. Sampling was carried out between September 2005 and October 2007.

Results and discussion

Summarized statistical data of the analyzed water samples are given in Table 1. The agricultural wells had the highest mean nitrate values of all the analyzed groundwater bodies (Table 1). In contrast, phosphate values are greatest in the

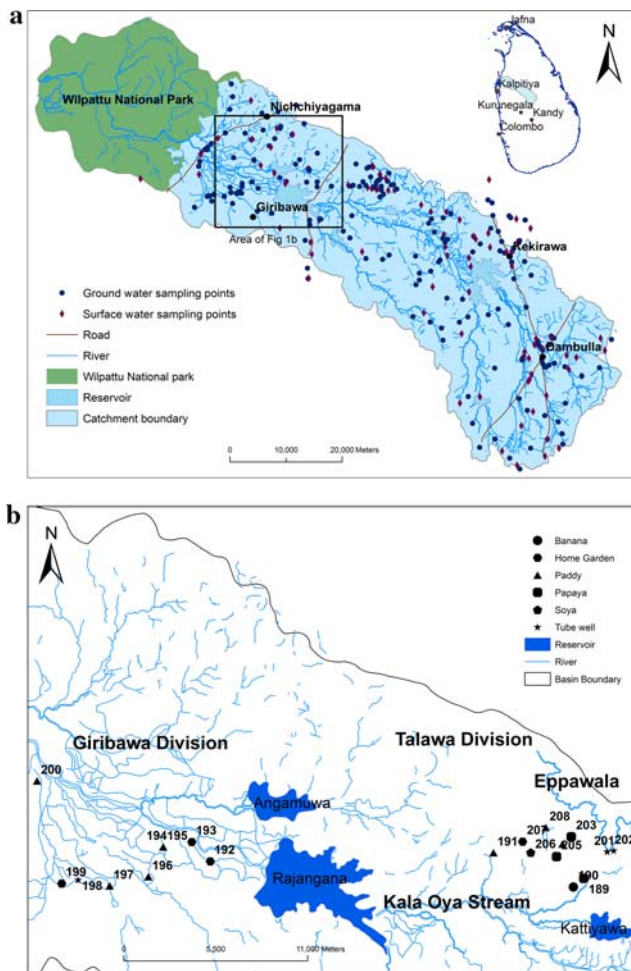


Fig. 1 **a** Map showing sampling points and location of the study area within Sri Lanka. The darker area is a National Park (forest), which was not sampled due to access restrictions. **b** Map showing Giribawa and Talawa secretarial divisions located on the left and right bank of the Kala Oya stream, respectively, and sampling points selected for the temporal study

tube wells (Table 1). Table 1 shows that the lakes contain significantly higher concentrations of nitrates and phosphates compared to the groundwater bodies and the other surface waters. Dissolved cation concentrations of waters from tube wells are considerably greater than in all other water types.

Spatial variation of nutrients in water

Spatial variations of nitrate and phosphate concentrations in the surface waters and groundwater in the study area are shown in Fig. 2a–d. Higher nitrate concentrations occur in the surface waters in two distinct areas in the central and eastern parts of the study area (Fig. 2a). However, phosphate concentrations in the surface water bodies show little variation, although the single area where phosphate values

are a little higher corresponds with one of the two areas where nitrate values are also high (Fig. 2b). Concentrations of nitrate in the groundwaters generally show little spatial variation with the exception of several small isolated areas (Fig. 2c), whereas relatively high phosphate values occur over much of the study area (Fig. 2d). The nutrient distribution maps show that the concentrations of nutrients in the surface waters are not related to those in the groundwater.

Comparison of the nitrate and phosphate concentrations at individual locations shows a positive correlation, with differing clustering between the groundwater and surface water (Fig. 3a). In addition, comparison of the average values of phosphate and nitrate at the locations where temporal variation was examined (Talawa area) show a positive correlation with a Pearson correlation between average nitrate and average phosphate of +0.395 (P value = 0.085), yielding a regression equation of average nitrate = $0.935 + 0.403$ average phosphate (Fig. 3). This relationship suggests that fertilizer application to the agricultural land plays a major role in the hydrochemistry of the area, especially in the groundwater. The correlation between the average values of nutrients in the field area may be statistically low, but the effect of fertilizer application remains of concern.

Nutrient variation in the surface waters

Nitrate concentrations in the surface waters generally show little variation (below detection limits—2.0 mg/l) throughout the basin. However, a few lakes exhibit very high values ranging from 10 to 30.0 mg/l (Fig. 2a). These high values may be related to cattle effluent, which can give rise to elevated nutrient levels. Although the extremes exceed the WHO limit of 10 mg/l, most sample sites exhibit very low nitrate levels (below detection limits—2.0 mg/l). The nitrate values observed in the surface waters indicate the low nitrate accumulation rates, despite the high application rates of nitrate as urea fertilizer in the cultivated areas. The surface waters flow throughout the year, in volume. Consequently, the surface waters tend to have low concentrations of nutrients due to dilution.

Heavy growths of nitrogenous plants (e.g., *Ipomoea* spp., *Salvinia*) are present along the banks of the canals, even during periods of non-cultivation. This indicates that the riparian zones absorb nutrients from the waters, which carry nutrients from the cultivated areas. This process may also thus reduce the nutrient levels in the surface waters (Hill 1996; Lamontagne et al. 2001).

Phosphate concentrations in the surface waters (Fig. 2b) generally fall in a narrow range (0.01–1.0 mg/l), although slightly higher values were occurred at some sites (1.0–2.4 mg/l). More extreme phosphate values (>21.7 mg/l) were observed in some lakes where advanced eutrophication was evident. These extreme values may be due to

Table 1 Concentrations of dissolved ions and pH of dug wells, agricultural wells, tube wells, lakes, canals, and streams

Groundwater	Dug well, <i>n</i> = 107			Agricultural well, <i>n</i> = 37			Tube well, <i>n</i> = 47		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
Nitrate	1.45	BDL	25.3	1.96	BDL	30.8	1.79	BDL	12.5
Phosphate	0.52	0.03	1.95	0.36	0.05	1.43	0.56	0.02	1.19
Mn	1.77	BDL	2.51	0.24	0.02	1.43	0.14	BDL	0.79
Fe	0.2	BDL	1.59	0.38	BDL	2.11	0.21	BDL	1.39
Mg	33.32	1.22	139	25.46	6.84	103.6	36.12	1.81	173
Ca	45.23	1.01	442	20.57	1.79	79.4	60.18	2.75	624
K	4.5	0.3	80	3.27	0.35	15.3	3	0.23	12
Na	69.18	9.07	545	92.49	5.77	719	99.74	12.35	575
pH	7.2	4.8	10.3	7.5	6.6	8.8	6.8	5.8	7.5
Surface water	Lakes, <i>n</i> = 59			Canals, <i>n</i> = 27			Stream, <i>n</i> = 19		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
Nitrate	2.37	BDL	29.9	1.41	BDL	10.7	2.74	BDL	23.4
Phosphate	0.52	0.01	21.6	0.51	0.02	2.6	0.21	0.22	0.66
Mn	0.3	0.03	3.08	0.4	BDL	2.04	0.12	0.01	0.44
Fe	2.04	0.12	21.9	1.18	BDL	6.1	0.68	0.02	3.89
Mg	19.72	1.33	75.8	30.73	1.12	93.8	22.01	9.7	55.1
Ca	18.06	0.86	93.2	20.28	6.06	48.9	18.32	2.52	25
K	7.5	1.16	32.7	3.51	0.21	10	2.36	0.91	9.11
Na	54.74	2.4	548	31.64	7.05	100.3	17.49	4.73	60.7
pH	7.4	4.7	9.4	7.2	5.6	8.4	7.2	5.9	7.9

Concentrations of dissolved ions in mg/l except for pH BDL—below detection limit, for Nitrate 0.1 mg/l and for Fe and Mn 0.01 mg/l, respectively

accumulation of phosphates derived from agricultural fields located within the catchments of such lakes. Furthermore, the lakes are prone to rapid falls in water level during dry weather conditions. At such times lake levels may fall to as low as 10–15% of their capacity, leaving all livestock in the area relying on this restricted water source. The result is that livestock effluent also contributes to increased nutrient loads in the lakes. The lakes with highest phosphate levels are also heavily invaded by plants, especially *Ipomoea* spp. These fluctuating water levels prevail in most of the lakes that are mainly fed by rainwater, and continuous nutrient accumulation may thus pose a threat to the future use of these water bodies for agriculture. In contrast, the lakes that are fed by canals are relatively poor in phosphate, indicating that ions leached from agricultural areas are diluted by irrigated water. Although flow rates in some of the canals crossing the agricultural fields are low, phosphate concentrations within them are also low. These canal systems therefore exert a major influence on water quality, as shown by Diez et al. (2000) and Bohlke (2002).

In general, the surface waters of the entire study area have the same chemical quality in terms of all dissolved ions. This suggests that the irrigation water sent through the

canal system also maintains the chemical quality of the surface water throughout the area. When the water is released to the canals, the flow rate is high, and consequently the water usually does not exhibit much chemical variation over time. In the case of the canal system, the canals are inter-connected, and their water flows to almost all the agricultural fields in the area. Consequently, due to this type of canal network, the water in the canals has low nutrient concentrations and shows little variation throughout the study area.

Nutrient variation in the groundwater

In contrast to the surface waters, the nitrate levels in groundwater are highly variable (0–30.8 mg/l) with clusters of higher values (10–30.8 mg/l) in localized areas. However, most of the nitrate values are low and lie in a narrow range of 0.8–1.7 mg/l. A limited number of samples from agricultural wells located around heavily cultivated areas within the paddy fields had high nitrate contents (1.7–12.5 mg/l), as did some deep groundwater tube wells. This indicates that the movement of nitrate within the surface and deep aquifers may have been caused by external factors

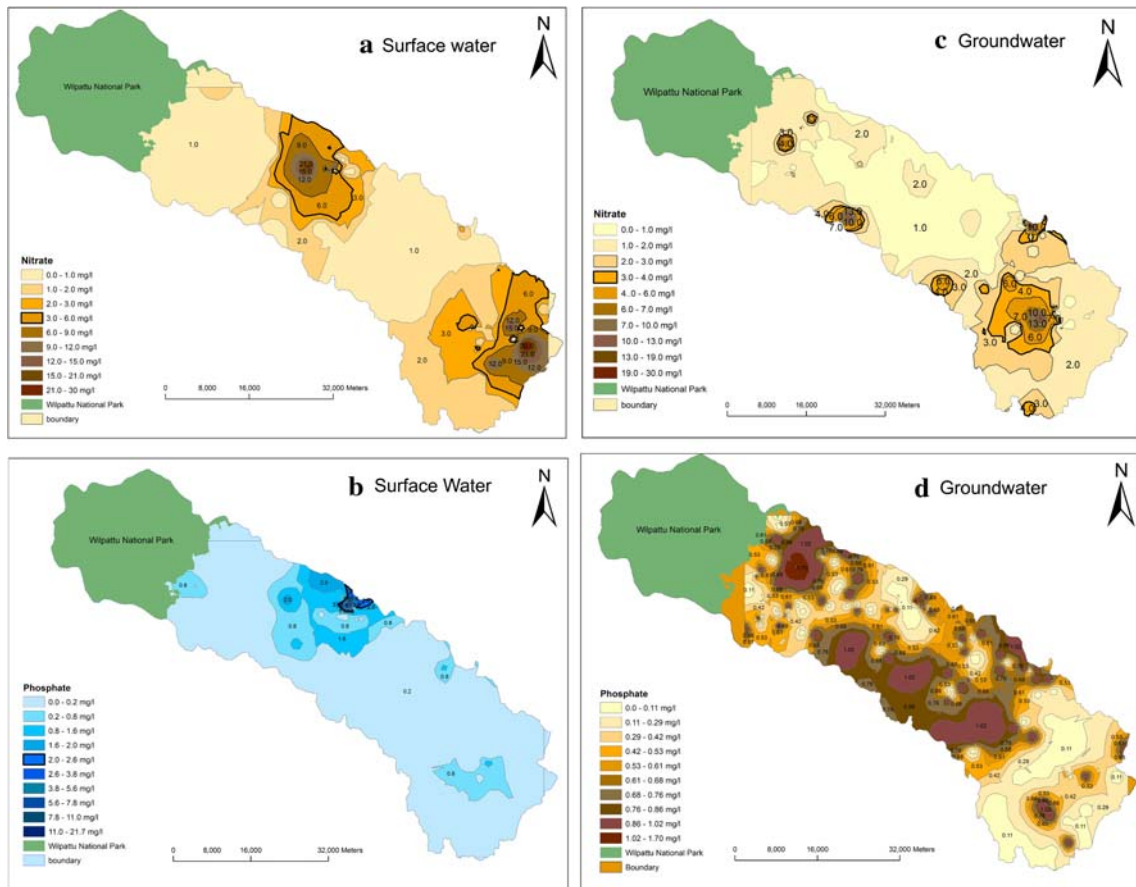


Fig. 2 a Distribution of nitrate concentrations in the surface waters of the study area. Note that there are some areas with relatively high values, whereas most of the district falls within the narrow range of BDL—2.0 mg/l. Values above 3 mg/l indicate anthropogenic effect. This has been highlighted by a dark contour to show the area with nitrate >3 mg/l. b Distribution of phosphate concentrations in the

surface waters. The World Health Organization (WHO) limit for phosphate in water is 2 mg/l, marked by the dark line shows the area above 2 mg/l. c Distribution of nitrate in the groundwater in the study area. The 3 mg/l contour is highlighted in a dark line to show the areas that have been anthropogenically affected. d Distribution of phosphate in the groundwaters. All values are well under WHO limits

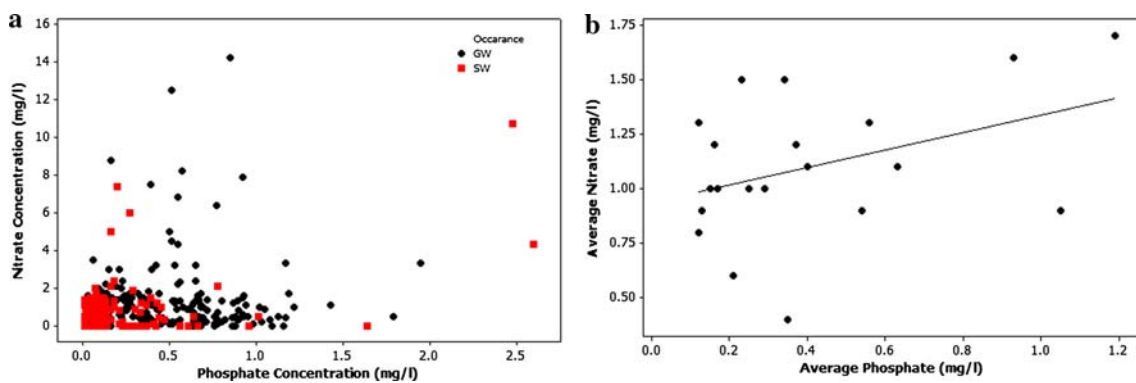


Fig. 3 a Average nitrate vs. phosphate concentrations of all data. b Plot of the average values of nitrate vs. phosphate concentrations of the locations studied for temporal variation (Talawa area). There is a positive correlation between the two ions (GW groundwater, SW surface water)

during the process of accumulation. Nitrate levels in groundwater of more than 3 mg/l are an indication of anthropogenic influence (Kite-Powell and Harding Anna 2006). Therefore, the high nitrate contents observed here

are an indication of groundwater contamination due to excess fertilizer application. Low concentrations of nitrate are prominent in the north-central district immediately beneath the areas in which most fertilizer applications are

made. Although fertilizer is applied continuously and at regular intervals, the groundwater is not yet vulnerable.

Phosphate concentrations in all the analyzed groundwaters fall in a narrow range (0.75–1.95 mg/l). The highest groundwater phosphate values are found around the Eppawala area (Fig. 2b) where a large phosphate deposit is located. The phosphate in the water may therefore originate from dissolution of rock phosphate. The results here show that the groundwaters generally have greater phosphate contents than the surface waters. This may indicate gradual accumulation of phosphate in the groundwaters.

Temporal variation

Temporal variation of nutrients in the groundwater at selected locations (Talawa and Giribawa, Fig. 4) shows a remarkable fluctuation following fertilizer application. However, values return to normal concentrations after 5 weeks.

The nitrate leachates from applied fertilizer take approximately 2 weeks to reach the water table in the area, and concentrations within the groundwater gradually decrease thereafter (Fig. 4a). In contrast, the phosphate variation is irregular (Fig. 4b) and values in the wells close to each other (within 1,000 m; e.g., paddy and soy data in Fig. 4b) show a similar variation with the time.

The variations in nitrate and phosphate concentrations in the well waters are similar irrespective of the type of agricultural practiced (Fig. 4). In contrast, nutrient concentrations in wells found in different soil formations show distinctive variations. During field investigations, we observed that sandy soils surround some paddy fields, and that the soils in these paddies were sand-rich. Wells that are situated in sandy soils surrounding paddy fields are

characterized by higher nutrient values than those situated in clay-rich soils (Torbert et al. 1999). This may reflect the low permeability of clayey soils (Addiscott and Whitmore 2003). Variations of this nature have also been observed in similar studies elsewhere (Bawatharani et al. 2004).

Gradual increase in the concentrations of nitrate and other solutes in the waters in the flow direction of the hydrological regime suggests that there is a greater threat of high contamination in groundwater at low elevations rather than at higher elevations (Fig. 5). This effect may become aggravated with time, due to accumulation of nutrients at lower elevations.

Factors causing low nutrient levels

As shown by Diez et al. (2000), Edwards et al. (1999), Lamontagne et al. (2001), Nolan (1999), and Rao (2006), nitrate and phosphate that reaches the groundwater may either be reduced, precipitated, transported, or adsorbed, depending on the conditions within the aquifer for each nutrient. Favorable conditions for decreasing nutrients are reducing environments, the texture and composition of the sediments, and higher concentrations of dissolved cations (Table 1) in the aquifer.

The comparatively low levels of nitrate in the waters analyzed here may be the result of a combination of factors noted in the study area. The main factor that contributes may be nitrate attenuation (Nolan 1999; Thayalakumaran et al. 2004) that results from the reducing conditions prevailing in the aquifers of the area. The data shows clearly that nitrate levels are low in Fe- and Mn-rich waters (regression fit, $Fe_{(mg/l)} = 0.342 + 1.562 Mn_{(mg/l)}$, P value = 0.000, Pearson correlation of $Fe_{(mg/l)}$ and $Mn_{(mg/l)} = 0.355$). Nitrate does not form insoluble minerals that could precipitate, nor

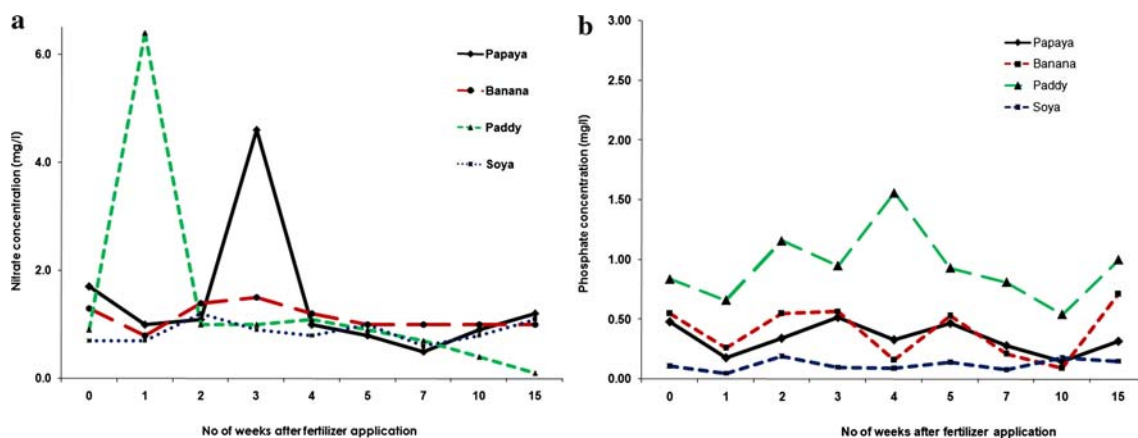


Fig. 4 Variation of nitrate and phosphate contents with time in selected agricultural wells in the Talawa area (from 18.05.2006 to 01.09.2006). **a** Nitrate. Concentrations are high at some sites the initial stage of fertilizer application for up to 4 weeks, and reach

background levels thereafter. **b** Phosphate. Application of phosphorus fertilizer does not significantly affect the phosphate values in well water

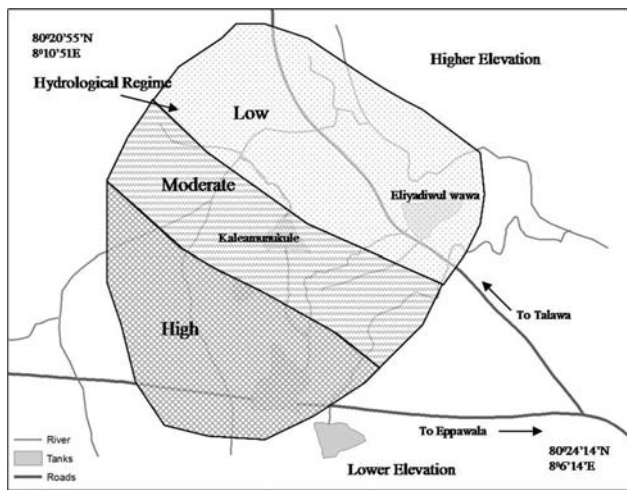
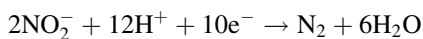


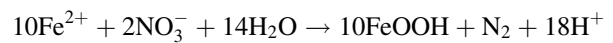
Fig. 5 Gradual increase of nitrate and other solutes toward the flow direction of the hydrological regime in the Giribawa area, showing contamination threat in the groundwater at low elevations Nitrate contents: Low 0.1–0.9 mg/l, Moderate: 0.9–1.0 mg/l, High 1.0–2.8 mg/l

is it absorbed significantly under aquifer conditions. The only means of nitrate removal from the system is by reduction. The necessary conditions required for reduction to occur are that the aquifer should be shallow, and that highly dissolved organic carbon should be present below the oxicline. These conditions are satisfied within the study area. Electron donors are necessary for the reduction of nitrate. Hence, the presence of high concentrations of cations in a reduced environment is evident. Under these conditions, nitrate can be reduced to N₂ by the reaction:



The reaction involves a complex pathway with intermediates of NO₂⁻, NO, and N₂O. Although kinetic problems can be expected, reduced groundwaters thermodynamically favor the reduction of NO₃⁻ to N₂. For substantial nitrate reduction in aquifers, reduction potential must be present within the sediments (Appelo and Postma 1993). The solid phases that can thermodynamically reduce nitrates are organic matter and Fe-silicates. Both are found in abundance in the study area, as also observed in a similar study by Bohlke (2002). Most of the sediments contain abundant organic matter, as confirmed by treatment with 30% H₂O₂. Organic matter thus very likely plays a major role in reducing nitrates in this district.

Iron-bearing minerals such as pyroxenes and amphiboles are common found in the basement rocks of the study area. Secondary iron-bearing phases should therefore be enriched in the overburden soils. Leached ions in such soil cover can also reduce nitrate to nitrogen (Thayalakumaran et al. 2004), by the reaction:



In addition, phosphate concentrations in the samples are also low in comparison to the fertilizer application rate. The higher concentrations of calcium and iron and negative correlation (Regression fit, Ca_(mg/l) = 206.5–17.46 phosphate_(mg/l), P value = 0.838) between Ca and phosphate (Fig. 6, inset) in the analyzed samples indicates that phosphate may be removed from the water through precipitation with cations at the presently available pH range, as shown by Spivakov et al. (1999). In addition, cation-rich humus soils contribute to the sorption process of phosphates (Angelo D’ 2005). When retention time is high, ionic interactions with the clay layers increase, causing precipitation, or sorption of phosphate from the water (Edwards et al. 1999; Bohlke 2002). These conditions occur in the study area, and may contribute to the reduction of phosphate in the waters.

The canals within the study area play a major role in controlling water chemistry by diluting the dissolved ions and forming reducing conditions. A large volume of water flows through the canals, and hence the dilution of nutrients is high throughout the basin. The canals are highly networked in a way that almost all the agricultural fields are fed with irrigated water. Consequently, the shallow groundwaters are regularly fed by the surface water, and the level of the water table increases abnormally. As a result, the chemical quality of the groundwater is controlled by the surface water, due to dilution and by production of the reducing conditions. The results obtained here show that most of the agricultural wells are characterized by higher contents of Fe and Mn and lower nitrate than the deep groundwater. The chemistry of the analyzed waters thus reflects the occurrence of reducing conditions in the shallow groundwater aquifers (Mansfeldt 2004). However, the dissolved ionic concentrations may differ in

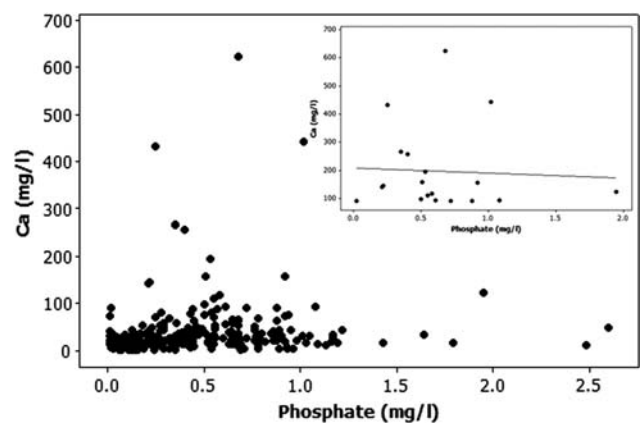


Fig. 6 The regression fit gives a negative correlation between Ca and phosphate, indicating that phosphate precipitation takes place in the presence of high Ca ion concentrations

each well due to difference in elevation, the size of the nearest canal, and the distance from a given well to the canal.

At high elevations in central Sri Lanka (Kandy area, Fig. 1a) the terrain is characterized by hill and valley topography. Although agricultural activity is not intensive in this area and other pollutant sources are comparatively low, very high nitrate values have been reported (Gunatilake and Gunatilake 2004; Rajakaruna et al. 2005). Sixty percent of shallow and open dug wells in the Kandy area contain more than 50 mg/l nitrate. This may be due to direct contamination through surface run off from high-elevation agricultural fields and from domestic waste. Similarly, very high nutrient concentrations also occur in water in the Jaffna and Kalpitiya districts, where soil permeability is very high (Dissanayake 1989; Liyanage et al. 2000; Rajasooriyar et al. 2002). In contrast, Silva De and Ayomi (2004) reported that the groundwater in the Kurunegala District, a flat terrain similar to our study area, contains low nutrient levels, with nitrate and phosphate levels of 0.1–0.08 and 0.21–0.98 mg/l, respectively. Although the water table in the Kurunegala area is deep, conditions are otherwise comparable with those in the north-central district. This may indicate that overburden soil conditions, man-made canal networks, and climatic conditions play a major role in the quality of the groundwater. This suggests that soil conditions and other external factors affect nutrient levels more than agricultural practices.

Conclusions

Nitrate and phosphate contents are low in the groundwater in the north-central district of Sri Lanka, despite high fertilizer applications. The main factor that affects nutrient levels is dilution by infiltration of water from the irrigation canals that distribute water to the agricultural fields of the area. This leads to relatively uniform water chemistry. Reducing conditions which develop in the irrigation water canals due to elevated water levels may also prevent the accumulation of nutrients in the groundwater. Iron-rich overburden soils in the area also have high potential to reduce the dissolved nutrients in the groundwater. Although nitrate contents in the waters are low, concentrations increase toward lower elevations. Since high nitrate values were observed in wells is the 2 weeks after fertilizer application, it is assumed that it takes approximately 2 weeks for leachates to reach the water table in the study area. Concentrations of nitrate and phosphate in the study area are lower than in other areas of Sri Lanka, and hence do not pose an immediate threat to the local groundwater resource. However, in the future the groundwater in the study area could be vulnerable to

contamination by leachates if excessive use of agricultural fertilizer continues.

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