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THEMATIC ISSUE

Factors that affect the spatial and temporal distribution of nitrate in a free aquifer of an agricultural plain basin

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Abstract

The behavior of nitrate in shallow aquifers depends on several factors, such as geomorphology, soil type, thickness of the vadose zone, and its lithology, sampling period, among others. In the free aquifer of the basin of the La Ballenera creek in southeastern of Buenos Aires province, Argentina, nitrate varies in a range between 4 and 250 mg/L in samples taken in spring and winter. This basin has a large agricultural production where fertilizers are applied, and nitrates come from them. Statistical analysis of the data allow to make conclusion about the main factors that influence the variation of the nitrate content in aquifers, in this case are the lithology and the thickness of the vadose zone, related to the different transit times. The results obtained can be extrapolated to analogous basins, considering that in the spatial analysis of nitrates the heterogeneities of the system should be considered. The importance of characterizing a system with more than one sampling at several sufficiently spaced times is emphasized.

Keywords Groundwater · Vadose zone · Soil · Geomorphology · Natural background · Contamination

Introduction

Elevated nitrate concentrations in groundwater are undesirable and it has become an important environmental indicator of human impact (Edmunds and Shand 2008; Sutton et al. 2011; Liu et al. 2015; Kim et al. 2015). The nitrate contamination of groundwater can cause risks to human health and the environment (L'hirondel and L'hirondel 2001; Powlson et al. 2008). The World Health Organization (WHO 2011) has set the threshold of 50 mg/L as NO_3^- for drinking water. Excess nitrate leads to methaemoglobinaemia (blue-baby syndrome) in infants less than 6 months old. Furthermore, it can pose health problems to pregnant women and gastric troubles in older adult, among other diseases (L'hirondel and L'hirondel 2001). Other researchers argue that there are other critical factors than the nitrate content in drinking water (Fewtrell 2004; Manassaram et al. 2010).

The *natural background range* and the *threshold* allow identified anomalous NO_3 concentrations in groundwater (Panno et al. 2006; Masetti et al. 2008; Giuliano Albo and Blarasin 2014; Cruz and Andrade 2015). Several researchers have indicated a relationship between agriculture and high nitrate concentration in groundwater (Jordan and Smith 2005; Liu et al. 2005; Burkart and Stoner 2008; Sutton et al. 2011, Wheeler et al. 2015; Amano et al. 2016; Valujeva 2016). Nitrate is the most stable of the inorganic nitrogen compounds in aerobic environments and is highly leachable through the soil profile into aquifers (Picone et al. 2003; Almasri 2007; Buczko et al. 2010; Baram et al. 2014). This is linked to low retardation coefficient relative to groundwater flow, low chemical degradation and diffuse continue character over time of the main sources of pollution (Foster et al. 2002; Chowdary et al. 2005; Rivett et al. 2006).

The content of nitrate in agro-ecosystems aquifers is affected by several factors: type, intensity and frequency of agricultural activities, soil type, climate variability, thickness and lithology of the vadose zone (VZ), and the own

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aquifer characteristics (Óenema et al. 1998; Hudak 2000; Lake et al. 2003; Muñoz et al. 2004; Rimski-Korsakov et al. 2004; Andrade and Stigter 2009; Brenes et al. 2011; Giuliano Albo and Blarasin 2014; Woli et al. 2016).

The main goal is studied the factors which contribute to the occurrence of high nitrate concentration in a typical agricultural catchment in humid plain area.

Study area

The study area is part of the Wet Pampa Plain and belongs to the geographical region Interserrana Plain which is extended between Tandilia and Ventania mountain ranges in the Buenos Aires Province (Argentina). La Ballenera is a small basin of 160.13 Km^2 with low slope around 0.6% (Calvi et al. 2016a) and orientation N–S perpendicular to the coastline. This stream flows from the Tandilia system to the Argentine Sea, but before that it feed La Ballenera pond near to the outlet (Fig. 1).The main towns in the catchment are Miramar and Comandante Nicanor Otamendi with 30,000 and 7000 inhabitants, respectively.

The climate has a ''moderate-humid'' climate (Köppen's classification) or ''sub humid–humid, mesothermal, without water deficiency'' type (Thornthwaite's method) (Thornthwaite 1948) with a mean annual temperature of 13.5 °C (CHEM 2013). In the period 1971-2015, the average annual rainfall in the catchment is 900 mm, and the average monthly rainfall is 74.9 mm (Calvi et al. 2014). Low slopes feature allows vertical processes, i.e., evaporation and infiltration to become dominant (Calvi et al. 2016a). During autumn–winter, the precipitation broadly exceeds the evapotranspiration generating an excess which mainly corresponds to infiltration (Calvi and Martinez 2018). The dominant vegetation was originally the grass steppe (Mosciaro and Dimuro 2009), but the region has been greatly altered by human activities, such as livestock and agriculture. At present the intensive agriculture is the major economic activity, mainly the crops of wheat, barley, maize, sunflower, soybean, and potato cultivation (Huarte and Capezio 2013).

Geology and hydrogeology setting

Several researchers have studied the regional geological and hydrogeological characteristics in the area (Sala 1975; Sala et al. 1983; Kruse 1986 and González 2005). The hydrogeological basement is included in "Complejo Buenos Aires" and "Balcarce" Formations. The multi-layered phreatic-semiconfined aquifer sequence is contained by layers of loess-like silt and silty-sand with precipitated CaCO$_3$ called "Pampean sediments¨ (Fig. 2). The mineralogical composition of these sediments was described by Teruggi (1954) and Martinez and Osterrieth (2013), and is mainly formed by quartz, plagioclases, and orthoclase with variable amounts of volcanic glass shards. The thickness of these Quaternary deposits varies between 30 and 100 m (Fig. 2) with typical hydraulic conductivity 10 m/d (Sala 1975), porosity of 15% and transmissivity around 800–1000 m^2/d. The groundwater is sodium bicarbonate type with pH around 7.3 (Calvi et al. 2016b).

Characteristics of geomorphology and soils

The study area is classified into two main morphodynamic units: system of hills and plain system plus shorelines (Calvi et al. 2016a). The first one has slopes from 1.71 to 15.8% and is formed by silt and silt-sandy composition sediments, with frequent calcretes (locally called tosca) interbedded (Fig. 1).

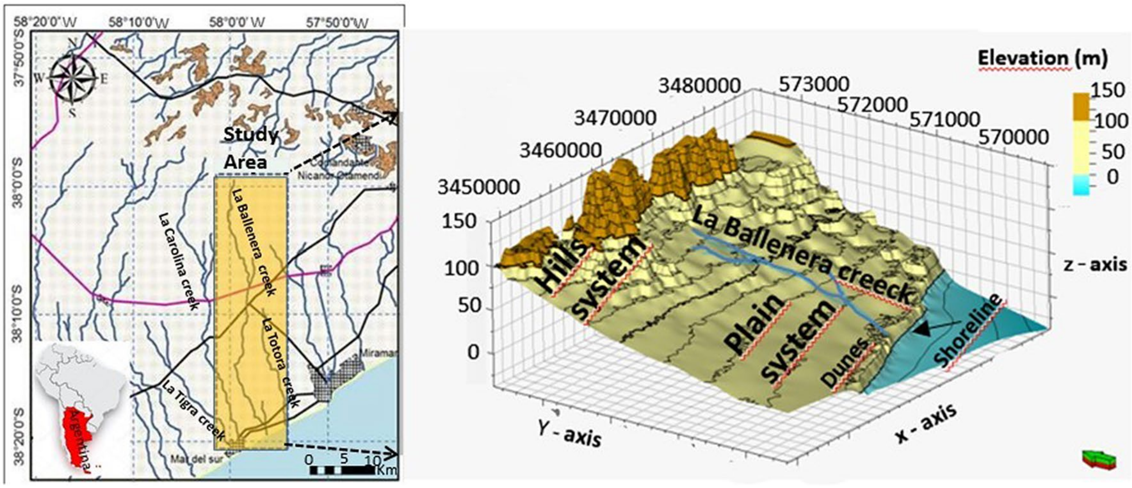

Fig. 1 Location map of the study area and 3D view of La Ballenera catchment

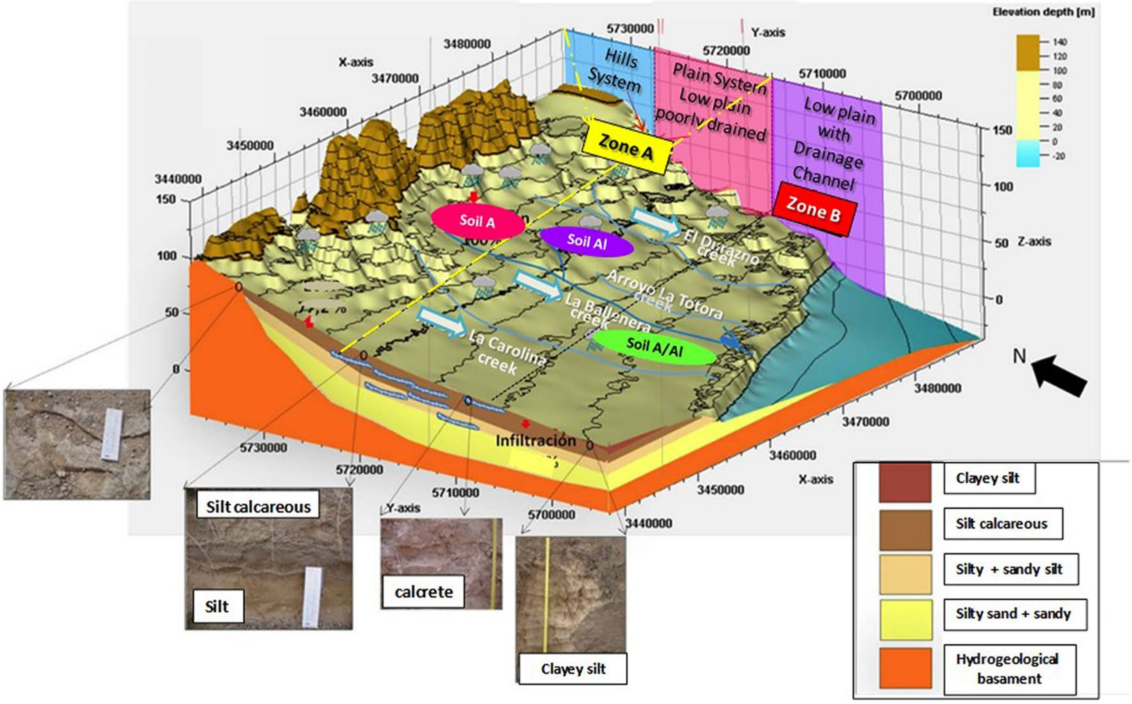

Fig. 2 Diagram block showing geomorphology and geology of the study area

The second one represents the largest plain in the basin and comprises a plain area slopes lower than 1.7% and a band of dunes next to the shoreline. The composition of this system is silt, silt-claying, and silt-sandy (Fig. 1) and two geomorphological units are distinguished: the poorly drained low plain (zone A) and the drainage drained low plain (zone B) (Fig. 2). The vertical hydrologic (infiltration and evaporation) are the dominant processes owing most of the basin has low slopes.

In the Interserrana area the predominant soils belong to the Mollisol Order (INTA 1989, 2011). These soils are the result of the action of a humid climate on loessic materials. At the head of the La Ballenera catchment, with slopes over 5.2%, of the dominant order is Argiudolls (A), with several subgroups of which the most important are the typical Argiudolls and Pretrocalcic Argiudolls (Fig. 2). The Argiacuolls soils (Al) are the main soils in the middle part of the basin with slopes lower than 5.2%. The soils next to the shoreline with low slopes of about 1.7% are Argiudolls and Argiacuolls (A and Al) (Fig. 2). These soils have 5–7% organic matter and 25–26% clay in the surface horizon (Costa 1999; Suero et al. 2002).

The main problems affecting these soils are sodium excess, impeded drainage, and high pH. However, the soils are generally well developed and widely distributed. They have dark colors and high contents of organic matter and nutrients. Those characteristics define them as the most fertile soil in the country (Álvarez et al. 2008).

In La Ballenera catchment, the *natural background range* statistically calculated is 4–11 mg/L (Calvi et al. 2018). The values above the threshold of nitrate calculated for the whole basin (> 11 mg/L) are related to agricultural activities introducing nitrogen in fertilizers and promoting the nitrate leaching which causes groundwater contamination.

Methodology

The sampling network for nitrate analyses consisted of 75 wells along La Ballenera catchment. These groundwater samples were taken on shallow windmill boreholes about 25 m depth, and irrigation wells about 50 m depth. A first groundwater sampling campaign for chemical analysis was carried out during October–November 2013 (spring season), when fertilization was being done. A second sampling campaign was followed up during July 2014 (winter season) without fertilization.

All sampling points were located by Global Positioning System (GPS) Garmin eTrex Vista for later use in maps and graphics. Also, water depth below surface level was measured in 56 wells, using a bipolar electric probe. The Geographic Information System (GIS) ArcGis 10.1 (ESRI 2012) was used to draw the piezometric lines and equal thickness maps. To achieve the objective, nitrate distribution maps were analyzed along the catchment in spring and winter campaigns. Nitrate analyzes were done by UV spectrometry in the Hydrochemistry and Isotope Laboratory of the Hydrogeology Group (University of Mar del Plata). The detection limit was 0.1 mg/L and the mean error was 1%.

Statistical analysis

The nitrate distribution was analyzed in relation to several factors as geomorphology, sampling depth (25 or 50 m), type of soils, thickness of VZ, and spring or winter crops. Some statistical parameters were obtained: mean nitrate concentration, standard error; standard deviation; number of analyses with nitrate concentrations higher than 50 mg/L with respect to the total number of samples and its percentage.

In addition, the box plots were performed to compare the medians of nitrate concentration between winter and spring sampling campaigns respect to the factor most representative. Finally, a diagram of NO_3 and VZ, was analyses according to the outcrop N–S of the catchment.

Results

Groundwater affected by nitrate

The nitrate distribution was analyzed according to the variables of geomorphology, sampling depth, type of soils, thickness of VZ, and seasons (spring or winter samples) (Table 1). The threshold 50 mg/L of nitrate is exceeded mostly to the geomorphologic plain area (zone B; 54.5% of samples) with average values of 83.4 ± 7.6 mg/L. Water is considered nondrinking when it has more than 50 mg/L nitrate, from the WHO regulations.

Respecting to the relationship among concentration and sampling depth, the first 25 m (windmills; mean of 64.5 ± 9.5 mg/L) show 57.5% of the samples that exceed threshold set by WHO (Table 1). The highest concentrations were detected in A/Al and Al soil type which have 83.2 ± 7.0 mg/L y 63.4 ± 8.8 mg/L nitrate, respectively, with 50.0 and 52.9% of samples exceeding 50 mg/L. Samples located in VZ with thickness < 2 m have a mean of 90 ± 7.2 mg/L and this value belong to zone B of the plain area, where 60% of samples exceed the limit established by WHO (Table 1). Finally, the spring and winter sampling campaigns have a similar nitrate concentration (Table 1).

The previous data show that the VZ is the most sensitivity variable responsible of nitrate content (60% of samples > 50 mg/L; Table 1). To demonstrate this result, box plots are performed to compare in both samplings campaigns the medians of nitrate concentration respect to VZ thickness. Also, Fig. 3 shows the thickness of VZ in the basin. The highest nitrate concentration is found in VZ < 2 m during spring. However, VZ from 2 to 6 m has higher nitrate concentration in winter than in spring (Fig. 3a).

In addition, Fig. 4 represents a dispersion plot of VZ thickness vs. nitrate content differentiating spring and winter samplings. In VZ < 2 m where the lithology is mainly silt, Fig. 4 shows that high nitrate concentration are grouped in spring sampling and low nitrate concentration are clustered in winter sampling. However, VZ 2–12 m is formed by thick calcrete bank and hardy silt with carbonate content, showing different behavior in the spring and winter nitrate value (Fig. 4). The lithology of VZ > 12 m is silt and calcrete, in spring and winter sampling exhibit low nitrate concentration.

The nitrate concentration was analyzed in space and time along the catchment through 3D diagrams and maps (Figs. 5 and 6). The highest concentration of nitrate is next to the discharge in October (spring) (Fig. 5). However, the highest values in winter (July) are in the middle of the basin (Fig. 6).

Table 1 Mean Nitrate (mg/L); Standard Error (SE); Standard Deviation (SD); Number of analyses in spring with $NO_3 > 50$ mg/L respect to the total samples (Num); ≅% of samples with $NO_3 > 50$ mg/L

Variable	Subcategory	Mean	SE	SD	Num.	%
Geomorphology	Zone A (*n* = 53)	51.6	10.0	2.7	23/53	43.4
	Zone B (*n* = 22)	83.4	7.6	2.9	12/22	54.5
Death	~ 25 m (*n* = 48)	64.5	9.5	2.7	22/48	57.5
	~ 50 m (*n* = 27)	54.5	7.8	2.6	12/27	44.4
Type of soils	A (*n* = 22)	39.9	9.1	2.8	7/22	22.5
	Al (*n* = 36)	63.4	8.8	2.8	16/36	50.0
	A/Al (*n* = 17)	83.2	7.0	2.8	9/17	52.9
VZ	< 2 (*n* = 20)	89.6	7.2	2.7	12/20	60.0
	2–6 (*n* = 321)	56.2	8.4	2.8	12/31	38.7
	> 6 (*n* = 24)	44.6	7.5	2.7	9/24	47.8
Samples	Spring (*n* = 34)	60.4	8.6	2.7	12/34	35.3
	Winter (*n* = 41)	61.4	9.2	2.8	19/41	46.3

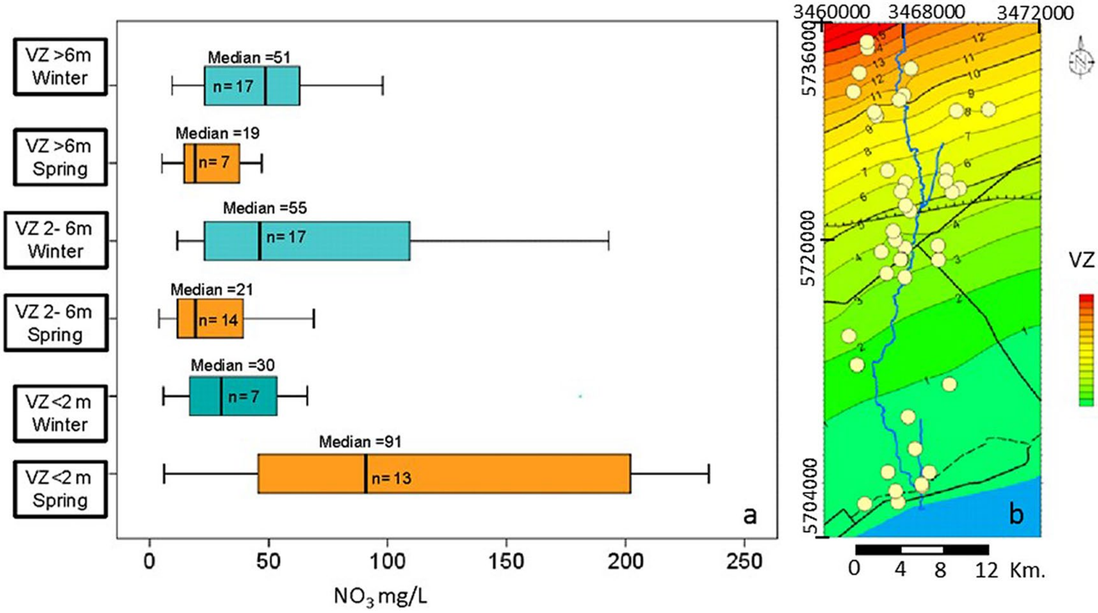

Fig. 3 **a** Box plots showing the distribution of groundwater nitrate concentration for different thickness classes of vadose zone; **b** Vadose zone thickness map

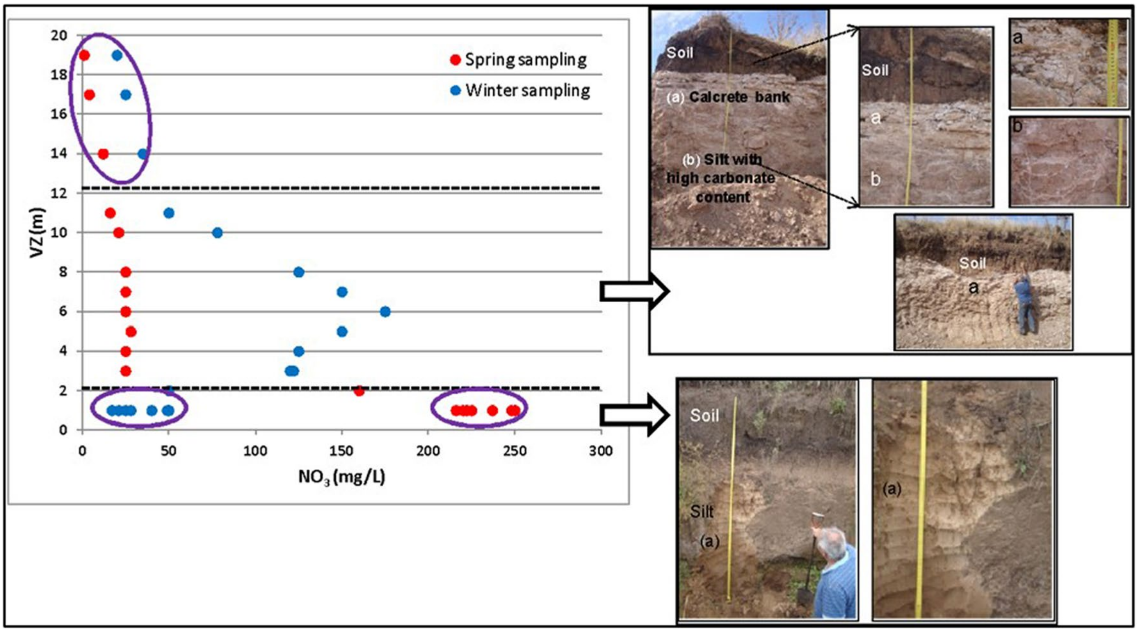

Fig. 4 Left: dispersion diagram N–S catchment between VZ and concentration of spring and winter sampling. Right: outcrops of the VZ 0–2 m and 2–12

Discussion

The high concentrations of nitrate observed (up to 250 mg/L) usually are related to agricultural activities as in other areas of the Pampean plain (Costa et al. 2002, 2011 among others) and the world (Follett and Hatfield, 2001; Wick et al. 2012; Hansen et al. 2016, 2017; Lawniczak et al. 2016; Qasemi et al. 2018; Kawagoshi et al. 2019). The addition of nitrogen fertilizer in crops, particularly in vegetables (potatoes, carrots, and beets) and wheat and barley (Echeverría and Ferrari 1993; Barbieri et al. 2009)

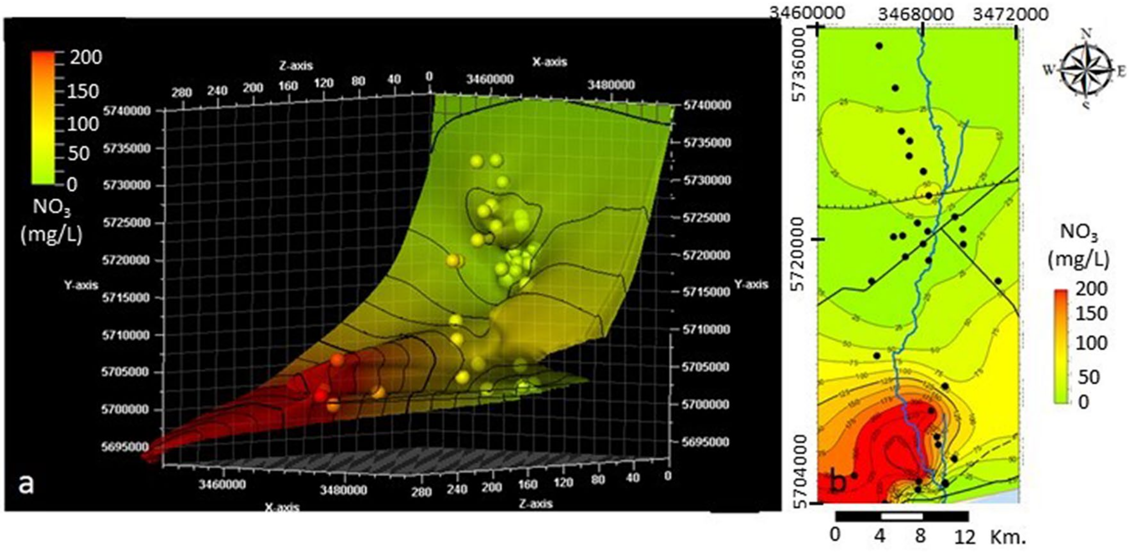

Fig. 5 Nitrate concentration: **a** Representation 3D in Spring (October); **b** Map of isoconcentration in Spring (October)

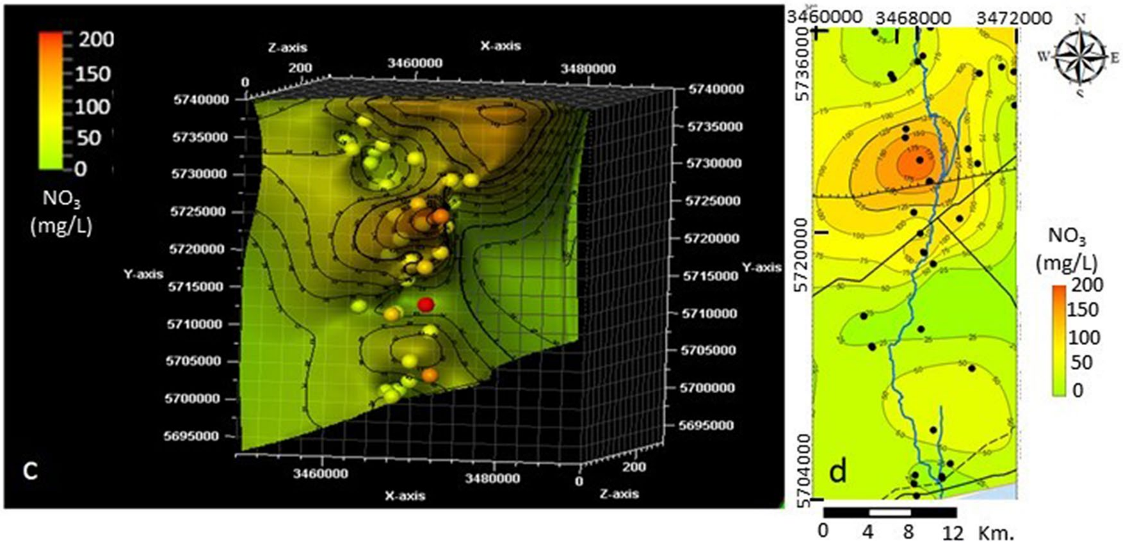

Fig. 6 Nitrate concentration: **c** Representation 3D in Winter (July). **d** Map of isoconcentration in Winter (July)

causes groundwater contamination. In the study zone, the period of fertilization goes from September to December when the probability of leaching is much lower because infiltration rates are lesser (evapotranspiration greater than precipitation). Nitrate leaching occurs when the soil is saturated with water and nitrate is washed beyond the root zone by percolating rainfall or irrigation.

The thickness and lithology of VZ are parameters that mainly impinge on groundwater vulnerability (Foster 1987; Blarasin et al. 2003; Fenton et al. 2009; Liu et al. 2015; among others). In the Ballenera basin, the zone A has the highest VZ thickness (mean: 8.5 m) and the lithology is composed by soil, silt, and discontinuous calcretes levels (Figs. 2 and 3). As well, the thickest calcrete bank was observed in the range of VZ 2–12 m and it has a moderate to high nitrate concentration in winter and lesser in spring.

In contrast, in the plain area with ponds (zone B) in the distal part of the basin, the thinner VZ (less than 2 m, Fig. 3b) has more sandy soils near coastal dunes and it has higher nitrate concentration in spring and lower in winter. Therefore, the nitrate concentration has opposite behavior during fertilized season (spring) and not fertilized season (winter) (Figs. 5 and 6). This result is linked to the different transit time in the VZ, which is lower in B than in A (mainly VZ 2–12 m), due to the lower thickness and lithology type (Fig. 4). As consequence, the nitrate value measured in Zone B is the result of the spring fertilization where the transport is dispersive. Whereas in zone A the nitrate applied in spring is measured in the next winter, showing successive pulses according to a piston flow model. Calvi and Martinez (2018) explained that the contrast in the isotopic composition of groundwater of these zones is consistent with the thickness and lithology type of both VZ related to different transit time. The hydrological balance confirms this assessment. The water excess calculated in winter (July 2014) allows the nitrate to be washed in the less thick area (zone B). The opposite occurred in spring (October 2013) due to the water deficit (Calvi et al. 2018).

These results show that performing nitrate concentration maps with a single sampling or two samplings separated by short times could be not adequate to show a realistic situation, because the arrival of nitrate to the upper water table can be driven by different mechanisms depending on the thickness of the VZ and the lithology. In the case of La Ballenera basin, where fertilization is applied in spring, it has been observed that for a sampling date it is possible to notice differences in concentrations in the low VZ area where a direct and recent impact is observed, against to the high VZ thickness where a continue and homogenized concentration is arriving.

Conclusions

The magnitude of nitrate leaching in the La Ballenera Catchment is influenced by geomorphology, type of soils, vadose zone thickness and its lithology, sampling season, among others, being lithology, and thickness of the VZ the main factors. It was determined applying a comparison of statistical analyzes and observing a map of the nitrate area from winter to spring samplings.

The results show different transit time and transport processes of the nitrate percolating from spring fertilization in vadose zone with variable thickness and lithology. Thus, it is necessary, on a side, to perform more than a sampling campaign to analyses the concentration of nitrate in phreatic aquifers. On the other side, it is demonstrated that analyzing all together the samples taken into a similar sampling period, the resulting distribution is not certainly consequence of the same fertilizer application season. The analysis of nitrate levels in an entire basin is a good practice if the heterogeneity of systems is also considered.

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