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Nitrate contamination of a rural aquifer and accumulation in the unsaturated zone

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Abstract

Groundwater contamination was studied in a rural setting of the Upper Pantanoso Stream Basin (UPSB) in the southeast of Buenos Aires Province, Argentina, where potential contaminant sources include inorganic fertilizer. Nitrate–N concentrations, greater than accepted level for safe drinking-water of $10\,\mathrm{mg\,l^{-1}}$ were present in 36% of sampled wells and 67% of samples had nitrate concentrations exceeding the background level of $5\,\mathrm{mg\,l^{-1}}$. Temporal fluctuation of nitrate concentrations in the groundwater was attributed to seasonal fluctuations in recharge and plant growth. Nitrate concentration was measured in deep soil profiles to determine the extent of leaching. Nitrate accumulation in the unsaturated zone of a soil cropped with potatoes was three times higher than the baseline N concentration found in the pasture. The greatest nitrate concentration in the soil profile occurred under irrigated corn where excessive nitrogen was applied. These results show that high fertilization rates and irrigation lead to increased hazards of groundwater pollution.

Keywords: Nitrate-leaching; Groundwater pollution

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1. Introduction

Public concern about groundwater degradation from point and non-point source contaminants continues to increase. Nitrate, found naturally at moderate concentration in many environments often, enriches to dangerous levels by human activity including the use of N fertilization.

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Nitrate ingestion with drinking-water by infants can cause low oxygen levels in the blood, a potentially fatal condition (Spalding and Exner, 1993). Moreover, nitrate concentration of 4 mg l^{-1} or more in water from community wells in Nebraska have been associated with increased risk of non-Hodgkin's lymphoma (Ward et al., 1996). Investigation conducted by local public health officials in Indiana implicated nitrate-contaminated drinking-water as the possible cause of several miscarriages (Schubert et al., 1997).

The US Environmental Protection Agency (USEPA, 1995) has established a drinking-water standard of 10 mg l^{-1} for NO_3^-–N. The Buenos Aires Province Law has established a standard for drinking-water of 50 mg l^{-1} for NO_3^- (11.3 mg l^{-1} for NO_3^-–N).

The local residents in the rural areas of the Upper Pantanoso Stream Basin (UPSB) use domestic wells as a source of drinking-water, while the town of Balcarce (36,000 inhabitants) located in the center of the UPSB has a public drinking-water system. Three of the eight public supply wells in Balcarce have been closed because the exceeded 10 mg l^{-1} for NO_3^-–N (data not published).

The impact of farming practices on NO_3^-–N pollution in ground and surface waters has been studied worldwide (Hadas et al., 1999; Cambardella et al., 1999; Patni et al., 1998). The extent to which agriculture in the "Humid Pampas" region contributes to water quality deterioration is not fully known. Auge (1998) and Hernandez and Minghinelli (1995) reported nitrate concentration that exceed 10 mg l^{-1} NO_3^-–N in particular areas of Buenos Aires.

Nitrate leached beyond the crop root zone must pass through the unsaturated zone before entering the groundwater. A survey of nitrate accumulation in the unsaturated zone may provide information about the impact of different agricultural practices on nitrate leaching (Katupitiya et al., 1997).

The objectives of this work were to monitor rural wells for groundwater nitrate concentration and to quantify the level of nitrate accumulation in the unsaturated zone for soils under different agricultural practices in order to assess their influence on groundwater contamination with nitrogen.

2. Materials and methods

2.1. Study area

The UPSB is located in the geological province named "Sierras Septentrionales" in the southeast of the Buenos Aires Province of Argentina (37°45′–37°55′S and 58°10′–58°20′W) covering an area of 194 km^2 (Fig. 1). Average annual rainfall is 900 mm and groundwater recharge is 100 mm per year (Suero, 1988). The dominant clay minerals of the UPSB soils are illite and montmorillonite.

The impermeable hydrogeological basement of the section is formed by a Precambrian crystalline rock (gneisses and granites). The overlying eopaleozoic sedimentary sequence is composed of ortho-quartzytes from the Balcarce formation (Dalla Salda and Iniguez, 1979). Secondary porosity has developed as a consequence of three faulting systems affecting the sediments. As a result of a high angle failure in the center of the watershed, the basement penetrates below the cenozoic sedimentary sequence (silt and fine sand from

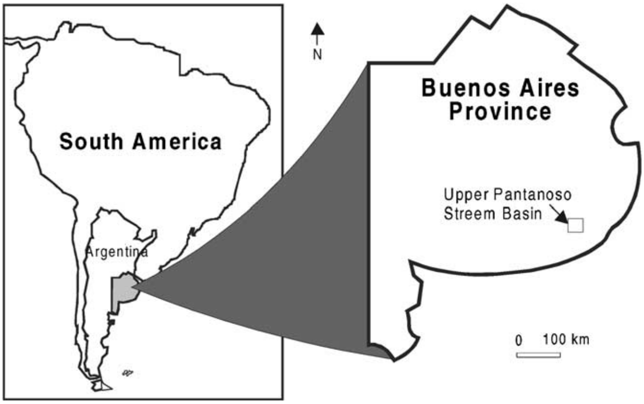

Fig. 1. Location map of the UPSB study area.

eolic or fluvial origin) that fill sectors of structural sinking and reaching thickness >100 m (Fig. 2).

The mineralogical composition of the aquifer is mainly quartz, plagioclase, potassium feldspar, and variable amount of amorphous silica in the form of volcanic glass (Teruggi, 1957). Occasionally micas and opaque minerals can be found. Variable proportions of calcium carbonate complete the composition of the aquifer.

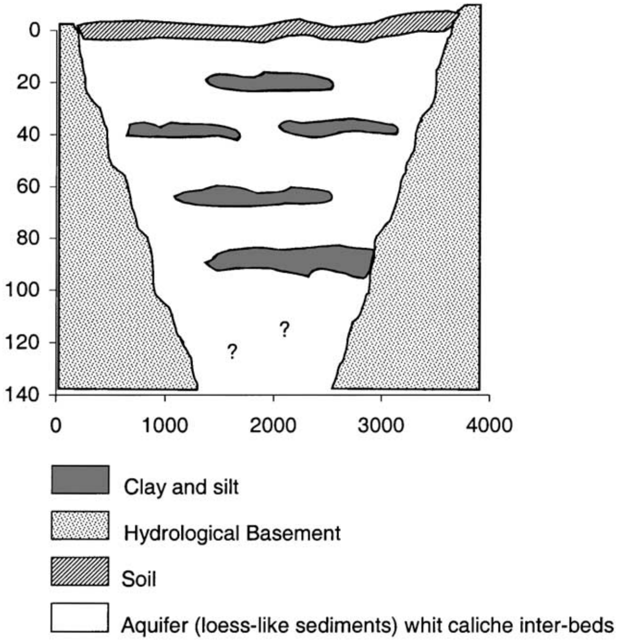

Fig. 2. Geological section of the UPSB study area.

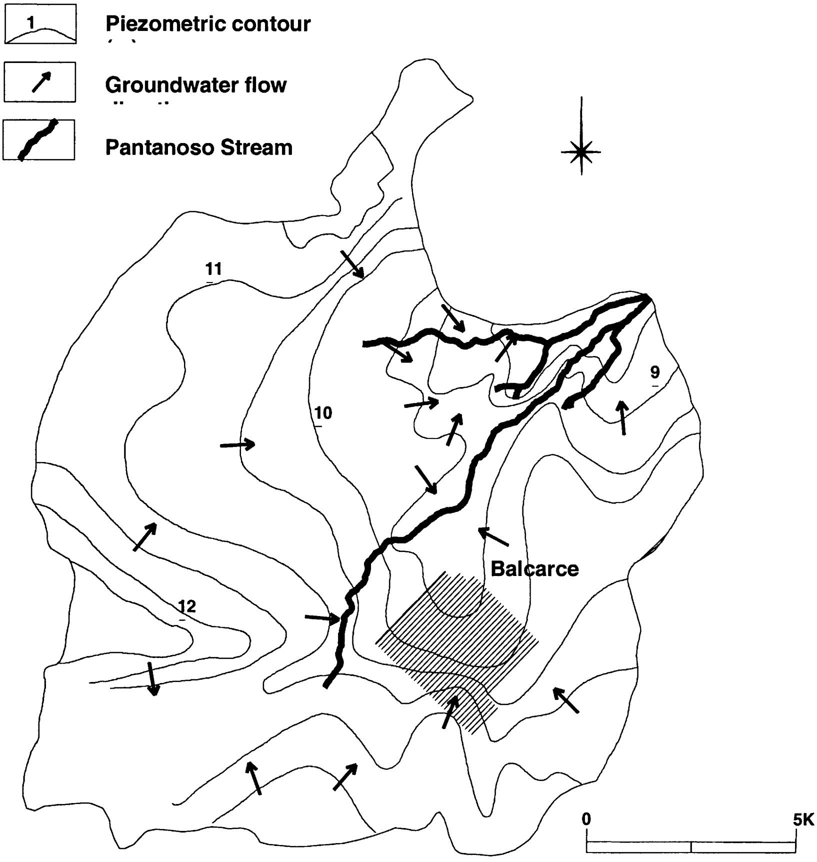

Fig. 3. Hydrological map of the UPSB study area.

Clay and carbonate layers with low permeability are inserted in the silt-sandy matrix. The hydro-geologic regime is a multi-layer free aquifer. The groundwater moves in the same direction as surface water from west-southwest to east-northeast (Fig. 3). Given the lack of artesian pressure of the aquifer, the recharge is produced by rain.

Water pH range from 7.5 to 8.0 and total dissolved solids varies from 500 mg l^{-1} in the recharge areas up to 2000 mg l^{-1} at the discharge areas of the basin due to progressive incorporation of dissolved salt along the flow path. At the boundaries of the basin, the water is bicarbonate—no dominant cation type, while at the discharge area the water is bicarbonate—sodium type.

2.2. *Soils and farm practicies*

The principal soils in the upper areas (croplands) of the UPSB are: Balcarce (fine–silty mixed thermic Paleustoll), Mar del Plata (fine–silty mixed thermic Argiudoll), and Cinco Cerros (fine illitic thermic Argiudoll) and the principal soils in the lower areas (grasslands)

Table 1
Some properties of soil within the area of UPSB

Soil	Depth (cm)	Organic C (%)	Sand[a] (%)	Clay[b] (%)	pH[c]
Balcarce	15	4.11	42.8	25.7	7
	30	2.64	44.9	27.8	7.4
	50	1.2	40.4	31.6	7.4
	70	0.64	35.7	29.3	7.8
Mar del Plata	8	4.03	41.1	23.1	5.9
	28	3.06	44	22.4	6.1
	31	1.74	40.1	23.6	6.3
	50	1.36	37.5	33.3	6.7
	70	0.58	34.2	31.4	7
Cinco Cerros	23	3.5	38.4	30.7	5.8
	45	1.53	33.3	41.8	6.5
Ayacucho	11	4.42	36	20.2	7
	24	2.52	36	19.4	7.8
	40	0.85	24.6	47.6	8.6
	50	0.48	26.3	37.8	9
	75	0.14	26.3	17.8	9.3
Chelforó	7	2.45	40.1	16.1	8.1
	30	0.98	26.5	35.5	9.5
	50	0.37	17.8	45.4	9.4
	70	0.24	26.3	26.5	9.2
Cobo	16	3.88	51	20.5	6.4
	41	3.72	46.4	23.3	6.3
	55	1.1	54.4	17.5	6.9
	78	0.79	40.9	39.1	7.4

[a] Walkley Black method.
[b] Textural analysis by the pipette method.
[c] Soil of 10 g and deionized water of 25 ml.

are: Cobo (fine mixed thermic Hapludoll), Ayacucho (fine illitic thermic Natraquoll), and Chelforó (fine illitic thermic Natraqualf). Soil properties are provided in Table 1 and a map shows the divisions between croplands and grasslands in Fig. 4.

In croplands, the farming practices consist of supplementary irrigated potatoes in a 5-year rotation, under conventional tillage. The rotation includes wheat, corn and sunflower. Many farmers grow pastures for cattle during 2 years before planting potatoes.

2.3. Monitoring studies

For this study, wind-mill wells were used to obtain water samples from groundwater. The mills were uniformly distributed in the area and pump water from 6 m below the groundwater table. Before sampling, the mills were pumped for 20 min in order to eliminate the stagnant water in the well. Water samples were collected in 1-l plastic bottles and stored at 4 °C prior to analysis. Two hundred and fourteen water samples were collected during February 1994, January 1995, October 1998, and October 1999. Variables

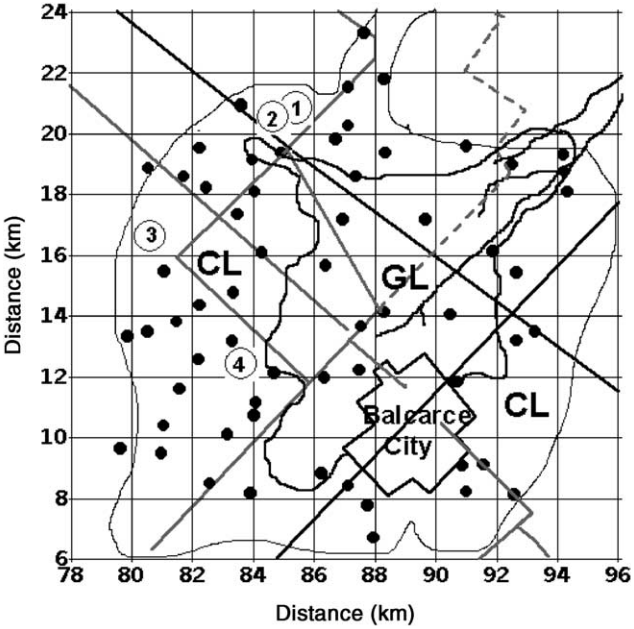

Fig. 4. Map of the UPSB study area showing, 1999 sampling points for ground water analysis (●); sampling profiles: (1) pasture, (2) corn, (3) and (4) potato; cropland areas under dash lines, and grassland areas outside of dash lines.

measured in water samples included pH, electrical conductivity (EC), Ca^{2+} and Mg^{2+} with atomic absorption spectrophotometer, Na^{+} and K^{+} with flame photometer, CO_3^{2-} and HCO_3^{-} by titration, Cl^{-} with specific electrode, and nitrate by steam distillation.

Deep soil samples (up to 5 m) were taken in duplicate with a drilling sampler from different sites: (i) two irrigated potato farms with 100 kg ha^{-1} of NO_3^{-}–N applied as urea at tuber initiation and 200 mm of irrigation during potato growing-season, (ii) an experimental plot where corn was grown with 200 kg ha^{-1} of NO_3^{-}–N applied as urea when the crop reach six leaves and 250 mm of irrigation during 5 years, and (iii) a plot under 5 years pasture were neither fertilization nor irrigation were applied. Soil samples were taken in 15-cm increments every 30 cm of depth and maintained at 4 °C until analysis. Gravimetric soil moisture was determined by drying part of the sample at 105 °C for 24 h.

Soil samples were analyzed for ammonium and nitrate. Both ammonium and nitrate were extracted from the soil with 4% K_2SO_4. Ammonia and nitrate were analyzed by steam distillation and titration with sulfuric acid (Bremner and Keeney, 1966).

3. Results and discussion

3.1. Nitrate in groundwater

The analyses of groundwater samples collected from several locations in the UPSB, indicated anomalously higher values of nitrate than the background. The background or

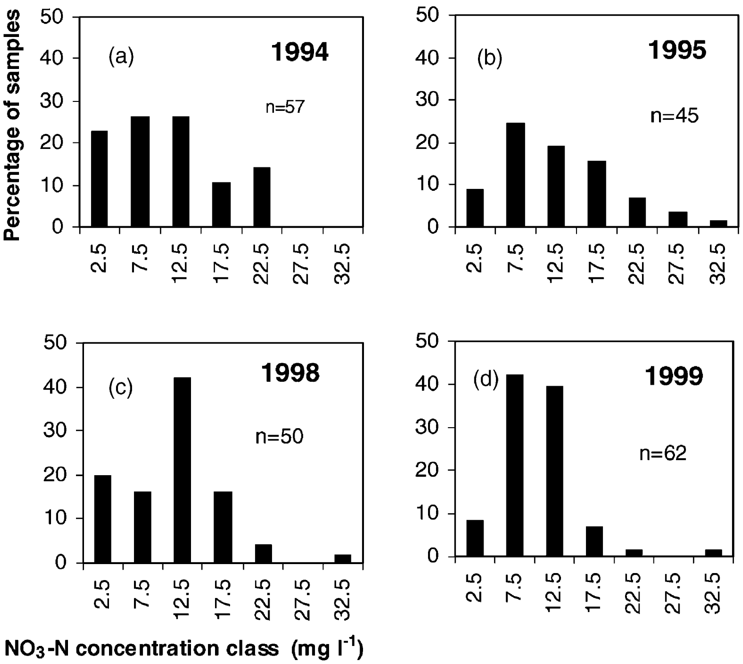

Fig. 5. Histograms of NO_3^-–N concentration distribution of groundwater sampled in (a) 1994, (b) 1995, (c) 1998, and (d) 1999.

natural value of NO_3^-–N groundwater content in the UPSB area ranged between 2.5 and 5.0 mg l^{-1} (Martinez, 1995). Mueller and Helsel (1996) reported that natural groundwater in USA typically contains <2 mg l^{-1} nitrate.

A large proportion of UPSB wells sample exceeded the 10 mg l^{-1} drinking-water standard NO_3^-–N, 33% (1994), 49% (1995), 33% (1998) and 26% (1999), while NH_4^+ were negligible. Histograms of the percentage distribution of NO_3^-–N concentration for different years showed different patterns of NO_3^-–N distribution (Fig. 5). The N concentration in the groundwater showed variations among years. For waters exceeding the drinking-water standard, the more frequent class was 10–15 mg l^{-1} and for waters under the drinking-water standard, 5–10 mg l^{-1} concentration was the higher frequency in 1994 and 0–5 mg l^{-1} class was more frequent in 1998. No temporal correlation between nitrate concentration in the same well was found. Due to the fact that nitrate concentrations in the wells varies over time, a single test provides limited information about whether the well is contaminated or not.

The spatial distribution of high nitrate concentrations appeared to be related with agricultural land-use patterns (Fig. 6). The areas with nitrate concentrations greater than 10 mg l^{-1} are located on the upper-parts of the basin where agricultural activity is more intensive (cropland), while the areas with NO_3^-–N values lower than 10 mg l^{-1} are located on the lower position where forage crops and pasture (grassland) are the predominant activities.

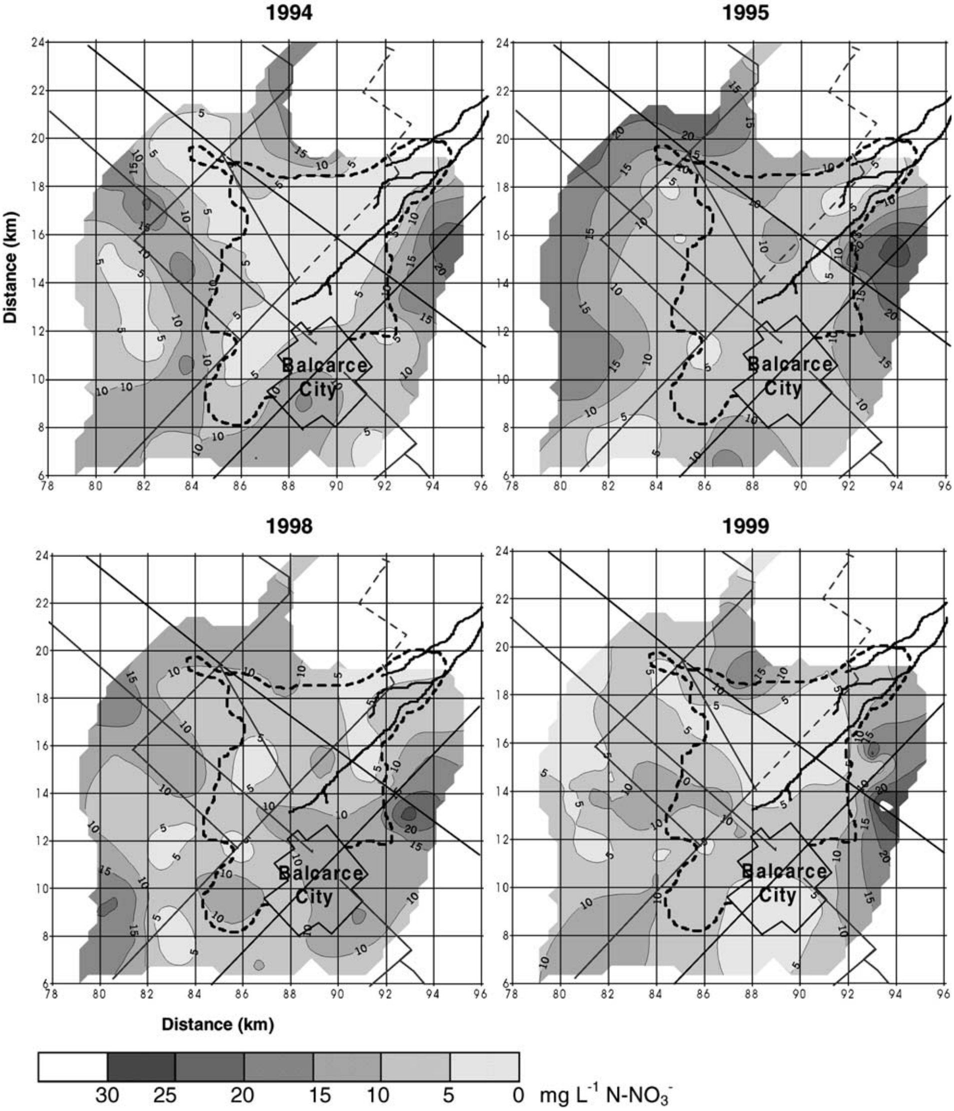

Fig. 6. Contour map of NO_3^-–N concentrations in the UPSB in 1994, 1995, 1998, and 1999 (grinding method: Krigging) showing: cropland areas under dash lines, and grassland areas outside of dash lines.

In the UPSB zone with higher proportions of croplands, between 30 and 69% of the domestic wells sampled had nitrate concentrations exceeding 10 mg l^{-1}, except for 1999. In the area under grasslands, this percentage ranged between 0 and 25% (Table 2).

The differential dilutions produced by variation in rainfall affected the spatial and temporal NO_3^-–N distribution. The low N concentration observed in 1999 coincided with a severe summer drought in 1998–1999 (Fig. 7). Temporal patterns in groundwater

Table 2
Percentage of wells with nitrate concentration >10 mg l^{-1} under cropped lands and for grassland at different dates

Land use	Percentage of wells			
	1994	1995	1998	1999
Cropped lands	59	69	55	30
Grassland	00	25	20	22

NO_3^-–N concentration in the UPSB depend on the asynchronisms of the processes described by Jaynes et al. (1999): N-uptake by plant, mineralization and precipitation.

While high nitrate water is present at the borders of the basin (croplands), high chloride water was found at the lower areas (grasslands) (Fig. 8). Chloride forms no complex with the soil and its salts are highly soluble. For these reasons, it is used as a tracer indicating the direction of the groundwater flow. Enrichment with NO_3^- reduces the $Cl^-:NO_3^-$ ratio (Weil et al., 1990). The high values of nitrate in the cropland groundwater indicates that agricultural activity is the main source of nitrate pollution. In this study, croplands have the lower $Cl^-:NO_3^-$ ratio supporting this inference. Land-use in the UPSB is closely related to water-table depth. Croplands are associated with a deep water-table (>4 m) and grasslands with a shallow one (<3 m). Lower NO_3^-–N values under shallow water-table conditions suggests denitrification, the process that converts nitrate to nitrogen gas which is fueled by organic matter in water-logged soils (Weil et al., 1990).

3.2. Nitrate in unsaturated zone

Nitrate accumulation in the unsaturated zone indicates N has been transported below the root zone (Fig. 9). The mass of NO_3^-–N in each 300-mm layer was based on NO_3^-–N

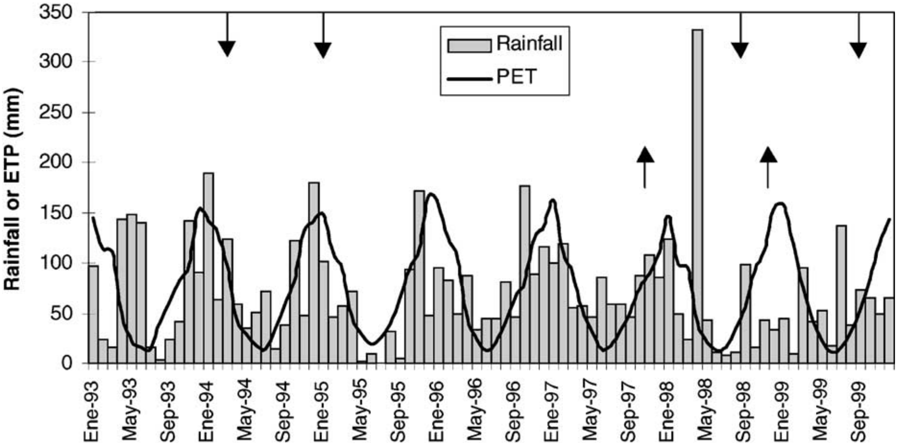

Fig. 7. The 3-month moving average rainfall and potential evapotranspiration (PET) from 1993 to 1999. Arrows pointing down indicate the date of water sampling and arrows pointing up the soil sampling dates.

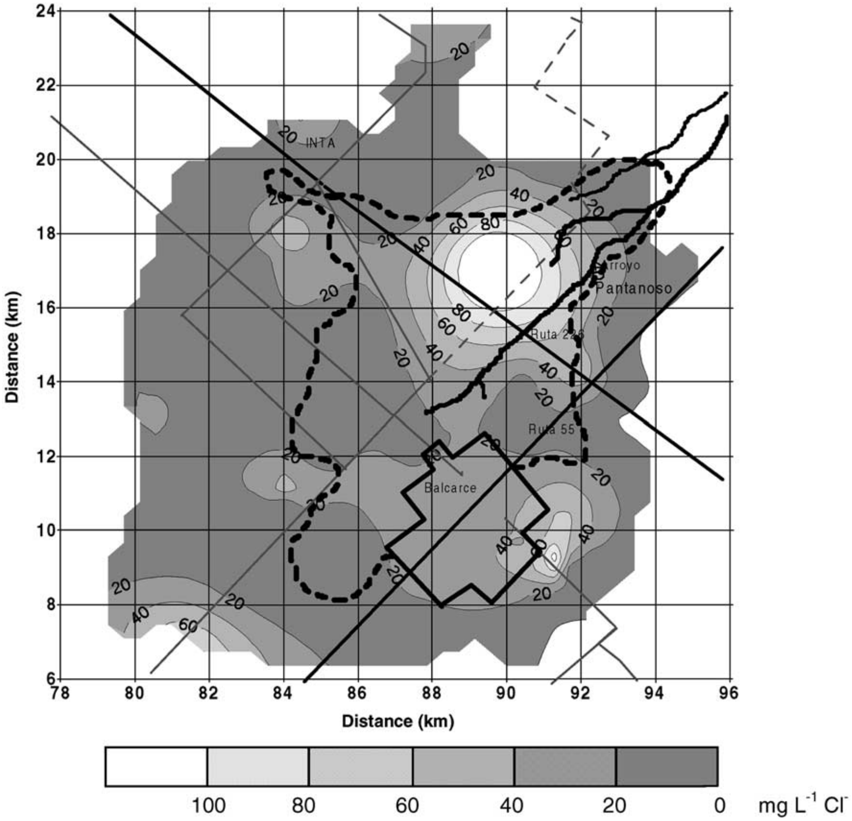

Fig. 8. Contour map of the Cl^- concentration in the UPSB in 1999 (grinding method: Krigging), showing: cropland areas under dash lines, and grassland areas outside of dash lines.

concentration, bulk density and the thickness of the layer. The water-table depth from most soils was below 6 m.

Soil NO_3^- beneath the plot with pasture was low, the profile did not have a defined peak of high NO_3^-–N and the amount generally was <5 kg ha^{-1}. The slightly higher level of nitrate in the bottom (3 m) came from the fertilization of the previous crops. Total nitrate amount stored in the 1.2–5.0 m unsaturated zone for the soil under pasture was 46 kg ha^{-1}, a value considered here as the baseline for the unsaturated zone.

A concentration of 260 kg ha^{-1} of NO_3^-–N in the unsaturated zone was found under corn receiving 200 kg/ha of N and 250 mm of irrigation. This amount is considered the potential amount of N that could be accumulated in the unsaturated zone; similar results were reported by Moreno et al. (1996). The amount of N present in the unsaturated zone of the potatoes farms (Fig. 9) was 160 kg ha^{-1} in 1997 and 156 kg ha^{-1} in 1998, representing more than 40% of the potential N stored in the unsaturated zone under irrigated corn.

Soil samples from potatoes and corn during spring 1997 showed an N peak at the 2.3-m depth (Fig. 9). However, the spring samples from the potato crop in 1998 showed an N peak

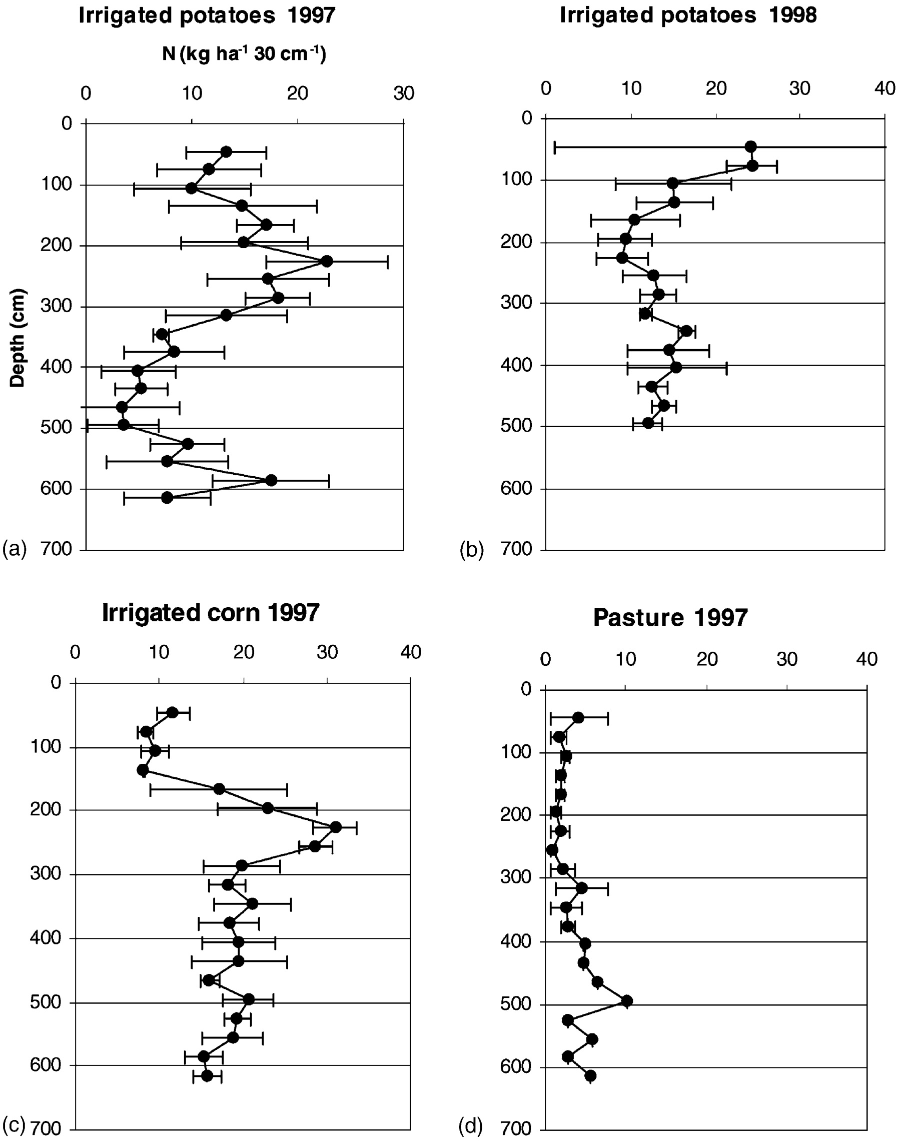

Fig. 9. Nitrate–N concentration profiles for: (a) irrigated potatoes (1997), (b) irrigated potatoes (1998), (c) irrigated corn, and (d) pasture. Errors bars indicate ± S.E.

at 3.5 m due to greater leaching from heavy rains during 1998 (Fig. 5). Deeper N peaks have less mass probably because of solute dispersion and pulse overlapping. Other common rotations (wheat, corn, sunflower without irrigation) presented 25 kg ha^{-1} of N in the unsaturated zone (Costa and Vidal, 1998).

NO_3^- in the unsaturated zone may leached into the groundwater depending on climatic conditions. Moreno et al. (1996) established that, in general, the NO_3^-–N content in the

Table 3
Estimation of the nitrogen available for N_l, N_p, N_m, N_f, N_d, and N_c

Crop[a]	Nitrogen available (kg ha^{-1})					
	N_p	N_m	N_f	N_d	N_c	N_l
Potato	50[b]	200[c]	100	6[d]	150[e]	195
Corn	55[f]	150[g]	200	6[d]	280[h]	140

[a] The NO_3^-–N budget was estimated using data from experimental information obtained in the INTA Experimental Station located in the UPSB.

[b] Saluzzo and Echeverria (1994).

[c] Echeverria and Bergonzi (1995).

[d] Sainz Rozas et al. (2001).

[e] Saluzzo et al. (1999).

[f] Echeverría and Sainz Rozas (2001).

[g] García (1996).

[h] Andrade et al. (2000).

soil profile at harvest time was leached during autumn and winter when the soil was bare. The patterns observed in Fig. 9 show a wavy vertical distribution of NO_3^- in the unsaturated zone. This distribution appears to reflect a combination of periodic pulses of water from rainfall and irrigation, and variations in NO_3^- availability due to crop uptake and mineralization.

The potential N available for leaching (N_l) at the end of growing season, for the irrigated potato and corn was estimated as follow:

$$N_l = N_p + N_m + N_f - N_c - N_d \quad (1)$$

where N_p is the soil NO_3^-–N at planting, N_m the NO_3^-–N mineralization in soil during the growing season, N_f the N applied as fertilizer, N_d the N denitrification, and N_c is the N removed by crop. The potential NO_3^-–N leached from the potato root zone according to Eq. (1) is 195 and 140 kg ha^{-1} for irrigated potato and corn, respectively (Table 3). These results are in line with the N concentration profiles suggesting the fact that irrigated potato could be a potential source of NO_3^-–N found in rural aquifers.

Mineralization may contribute a large proportion of the nitrate leached from arable land (Addiscott, 1988; Jenkinson and Parry, 1989). In the UPSB after potato harvest (February–June), temperatures are still high and mineralization of organic matter occurs. Tillage of the top 20 cm of the soil also contributed to mineralization after potato harvest. During fall and winter, when precipitation exceeded evapotranspiration, the accumulated NO_3^- is available to be leached.

3.3. *Nitrogen inputs from other sources*

Excreta returned to the sward, especially urine, create a ‘hot spot’ of high N content (Jarvis et al., 1989). From the annual N input of urine measured in the UPBS grassland (35 kg ha^{-1}, Videla et al. (1994)), some is volatilized as NH_3 (6.5 kg ha^{-1}, Videla et al. (1994)) and the N returned to the sward is 28.5 kg ha^{-1}. Some of this amount will be used by immediate local crop demands for N. At the grassland UPBS area, soils are frequently waterlogged and denitrification could take place, then part of the N also could be lost as N_2O. The amount of N available for leaching in this case could be then less than the estimated N value of 28.5 kg ha^{-1}. The low input of N from cattle and denitrification could be a reason of the low proportion of water samples exceeding 10 mg l^{-1} in grassland areas.

Another source of groundwater N contamination is sewage from septic systems. The population density in the rural areas of the Balcarce county is 0.1 person per ha (1991 population survey). The amount of N in human excreta is 5 kg per year (Foster et al., 1987) which results in an average input from domestic septic systems in the rural areas of 0.5 kg ha^{-1}. Besides, some rural areas have a population density larger than the average value used in this calculation, this potential pollution source is much lower than the pollution sources generated by agricultural activities.

4. Conclusions

Nitrogen fertilizers applied to fields and soil mineralization induced by agricultural activities appear to be the primary source of nitrate in the UPSB groundwater. Nitrogen not used by crops can be carried to the underlying aquifer. Irrigated agriculture is associated with high nitrate fertilization and high frequency of contamination of groundwater in the area under study. Since, most of the Buenos Aires province depends on groundwater for the domestic drinking-water, it would be desirable that alternative production techniques be tested to minimize nitrate–N leaching from agricultural fields.

Possible mitigation strategies that would reduce N accumulation in the unsaturated zone are: (1) to plant and incorporate a catch crop following potatoes harvest to immobilized inorganic N into organic pools; and (2) introduce deep-rooted crops like alfalfa in the rotation.

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