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"NPS pollution analysis in groundwater and streams of rural watersheds in western and southeastern Pampas, Argentina"

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Abstract.

Non-point source water pollution is a key question in rural watersheds and it needs to be studied in order to prevent damages to ground and surface water quality. The main goal of this study is to analyze nutrient and chemical loads in groundwater and streams in Pampa region, Argentina. For studying groundwater loads, a set of 19 observation wells were installed in 2011, in western Buenos Aires. The wells were located at three landscape positions (upper, middle and lower hill) in seven agricultural fields and groundwater samples were monthly collected. As for surface water, two watersheds located in southeastern Buenos Aires, were chosen: Napaleofu creek Watershed (34.000 hectares) and Quequen Grande River watershed (938.000 hectares). Daily water samples were taken from the main stream from October 2011 to May 2013, at both watersheds. Water Samples collected from wells and streams, were analyzed to determine N, and chemical loads. A group of 11 herbicides and one insecticide frequently used by farmers in the watershed were chosen for the study. Nitrogen and chemical concentrations were analyzed considering rainfall events and also compared to critical limits. Preliminary results are presented from a subset of samples since remaining samples are currently being processed in laboratory. As for NO_3-N concentration, most of wells presented variable concentration depending on monthly precipitation and landscape position. Considering 10 mg/L NO_3-N as a standard limit, 52% of the observations exceed this value mostly related to unusual precipitations events at winter 2012. Nitrate-N *concentration in streamflow at Quequen Grande River and Napaleofu creek were on average 5 ppm. NPS Pollution modeling is a second goal of this on-going research. SWAT validation results are also presented for one of the watersheds under study.*

Keywords. *NPS Pollution, Groundwater and surface water, Pesticides and nutrients, Agriculture, Argentina.*

Introduction and Objectives

In agricultural watersheds, variable amounts of pesticides can be released to streams and aquifers through surface runoff and leaching, jeopardizing sources of drinking water. On the other hand, pesticides make possible high agricultural yields. This process of chemical losses in rural areas is well known as Non-Point-Source Pollution or NPS Pollution. The driving force of NPS pollution is the rainfall-runoff process, which tends to be a complex non-linear, time-varying and spatially distributed process in agricultural watersheds.

Most of literature related to NPS pollution is mainly focused on nutrient losses, basically Nitrogen and Phosphorus, rather than pesticides (Dillaha et al., 1990). Nitrogen, as nitrate, moves in water solution by surface runoff, or vertically to groundwater by leaching. Phosphorus usually moves sorpted in sediments, so that is closely related to erosion processes. However, pesticides might also be transported to surface water bodies or to groundwater aquifers.

Even though rainfall-runoff process is the driving force for NPS pollution, the soil texture and the amount of chemical input to the system, are important variables to take into account. In sandy soils water movement is mainly vertical, increasing the risk of nutrient transport to groundwater. Conversely, in clay soils vertical water movement is very low and the risk of nutrients or chemical transport is mainly associated to surface runoff to streams or surface water bodies.

In Argentina, in the last years, the consumption of agricultural chemicals yearly increased since 1990’s. Figure 1 shows the evolution of the agricultural chemical consumption from 1997 to 2011 (Negri et al., 2009). This increment can be explained by two main drivers: expansion of agriculture and significant adoption of No Tillage system. For the period 1997-2011 crop area in Argentina increased around 28%, from 27 million hectares to approximately 35 million hectares. However, the increasing adoption of no tillage system was even more significant. In 2000, around 10 million hectares were cropped under no tillage, while in 2010 almost 26 million hectares were cultivated under this system (AAPRESID, 2012). . As known, tillage system preserves soil structure and improves soil carbon cycle, but it is more demanding in chemicals than conventional tillage.

For these reasons, a research was initiated in 2011 to study NPS Pollution in two areas of Pampa Region: Western Buenos Aires and Southeastern Buenos Aires. Western Buenos Aires presents somewhat excessively drained sandy soils under agriculture, making possible to study groundwater constituents at different farm fields. Southeastern Buenos Aires presents clay loam soils and a defined drainage pattern that make possible to analyze water quality on streams. At southeastern Buenos Aires two watersheds were chosen: Quequen Grande river and Napaleofu Creek watersheds.

The goals of this on-going research are two: (1) making a diagnostics about groundwater and surface water quality in representative rural areas under agriculture, in Pampa regions; (2) using all collected data to validate hydrological models, such as Drainmod –N II (Youseff et al., 2005), GLEAMS (Leonard et al., 1987) and SWAT model (Soil Water Assesment Tool) (Arnold et al.,1998). NPS Pollution modeling is considered for this research a key point, since mathematical models allow estimating environmental impacts for potential future scenarios, such as climate and landing use change.

This paper presents only some preliminary results that comes from subsets of water samples from all sites. It encompasses the whole period October 2011-April 2013. These samples were first analyzed for testing laboratory equipment and calibration curves. Most of groundwater samples were analyzed for nitrate concentration, but only 37 were finished for chemicals. As for surface water samples, a subset of 50 was analyzed for chemicals and nitrates. Most of results, including samples to be collected next months, will be ready at the end of the present year.

In addition, calibration and validation results for the hydrological components of the SWAT are presented. DRAINMOD model validation results have been presented as a separate paper at this ASABE 2013 International Conference.

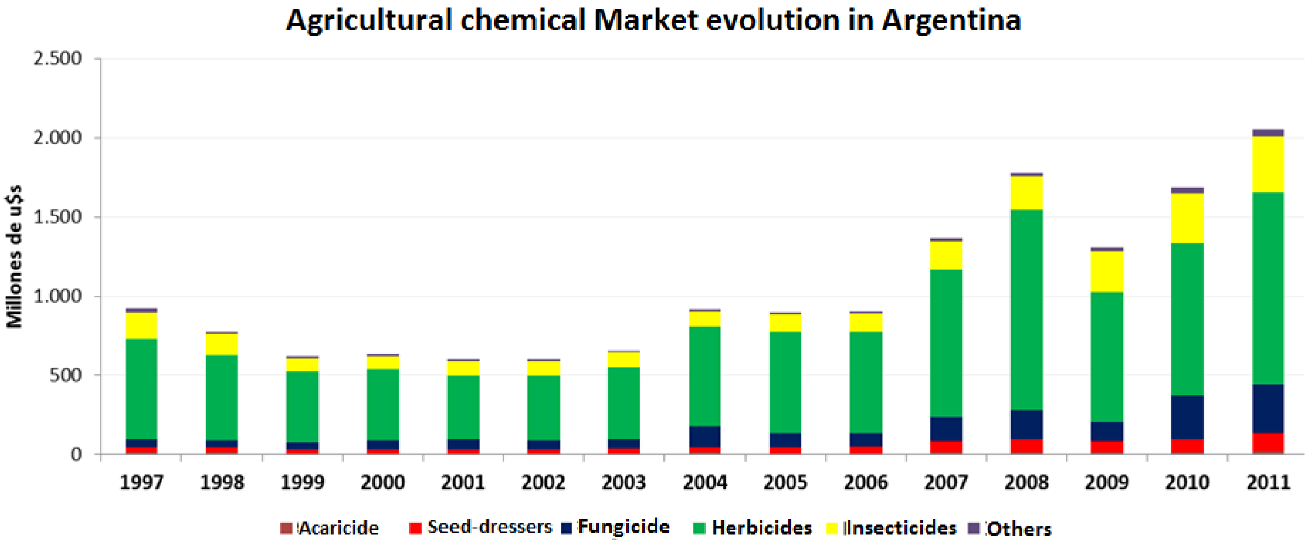

Figure 1 - Agricultural Chemical Market evolution in Argentina between 1997 and 2011 (Negri et al., 2009)

Sites description

This study was carried out at two different areas: Western and Southeastern Buenos Aires.

Western Buenos Aires

Western Buenos Aires has temperate climate and annual mean temperature is 16^0C. The average annual potential evapotranspiration approximates to 1250 mm. Mean annual precipitation depends on the period considered, but for the period 2000-2012 it was around 950 mm. However, there were differences among years with low records such as 655 mm/year, in 2005, and peaks like 1350 mm/year in 2002 that caused flooding. These variations have determined groundwater table depth depletion or increasing in relative short periods of time.

For this study, a set of 19 observation wells was installed in 2011, in western Buenos Aires. The wells were located at three landscape positions (upper, middle and lower hill) in seven agricultural fields. Groundwater samples were monthly collected (Figure 1).

These wells were located within crop fields to make sure that agricultural inputs would be applied on the shallow aquifer recharge area. Soils were deep and sandy Hapludolls, from “well drained” to “somewhat excessive well drained”. Groundwater table depth was monthly recorded and groundwater samples were monthly collected, refrigerated and submitted to laboratory, to determine nitrates and other constituents. Previous to take samples, the wells were pumped to remove remaining water. Water samples were taken after well recharging. Sampling period extended for 17 months, from November 2011 to March 2013. Nitrogen was expressed as Nitrate-Nitrogen (NO_3-N) to make it comparable with literature and US EPA standards.

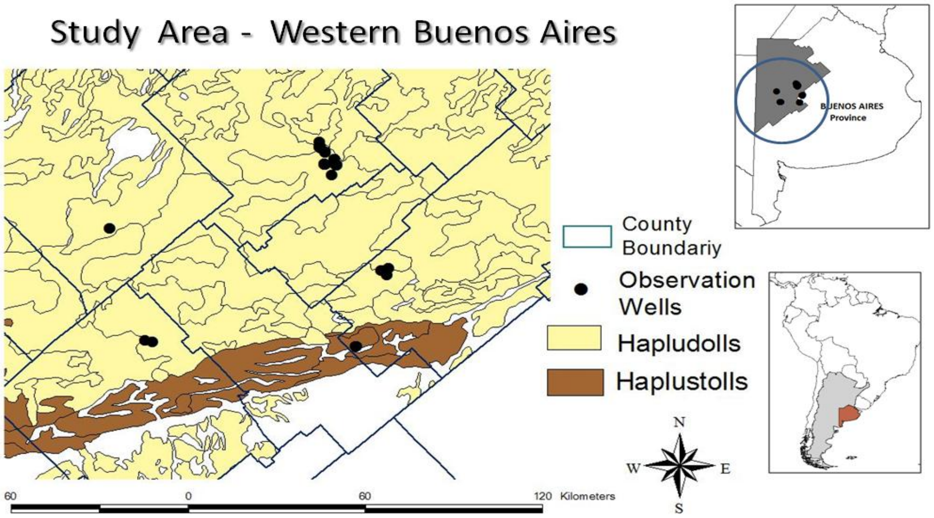

Figure 2 - Study Area – Western Buenos Aires Observation Wells

Southeastern Buenos Aires

In this area, the climate is temperate and the average annual precipitation approximates to 950 mm for the period 1970-2012. Soils are mainly Typic Argiudolls in upland and midslope sectors, and Natracuolls and Argiaucolls in poorly drained fields. Most of agricultural crop fields are under No Tillage system and terrace countours are frequently used, especially in the steepest lands of the Napaleofu creek Watershed.

As mentioned, two watersheds were selected at southeastern Buenos Aires province. Quequen Grande River watershed - which extends on 938.000 hectares -, and Napaleofu creek Watershed which occupies 34.532 hectares (Figures 2 and 3). Agricultural crops represent a 27% of the area of the Quequen Grande River watershed. The remaining 70% is occupied by pastures and natural grasses. The monthly average discharge of the watershed is around 16,5 m^3/s, but strongly depends on period considered. The average slope is approximately 1% or less.

Napaleofu Creek Watershed (Figure 3) is a smaller basin occupied almost 100% by agricultural crops. It is located northeast to the Quequen Grande river watershed. In both watersheds, winter crops represent around 50% of the total cropping area, and summer crops occupy the remaining area. This watershed has an average discharge of 0.4 m3/sec and the average slope is around 1.5%. However, it presents very steep sectors in the boundary with Quequen Grande river Watershed.

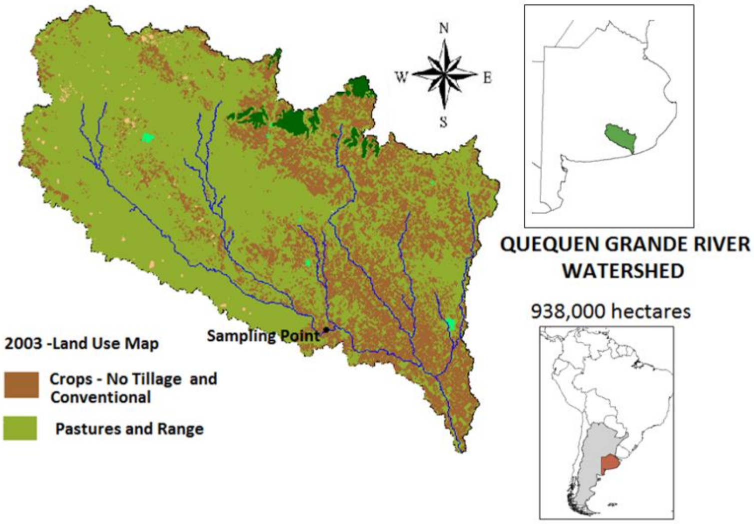

Figure 3 - Quequen Grande River Watershed

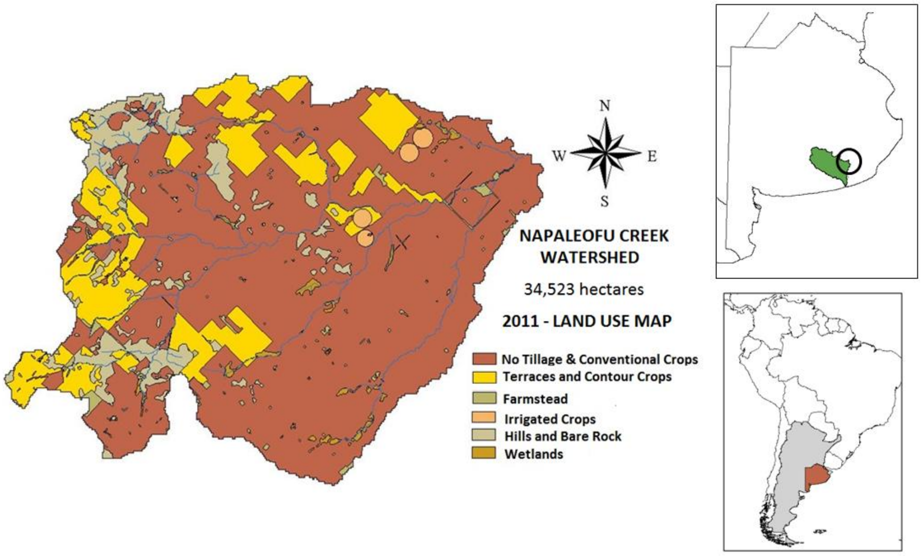

Figure 4 - Napaleofu Creek watershed

From October 2011 to April 2013, water samples were daily collected from Quequen River, at the sampling point located at “Puente Blanco” (Figure 2). Napaleofu creek water samples were taken daily, but only at week-days. All samples were refrigerated below 5 °C and sent to INTA laboratory.

Chemical Analysis

Previous to analyze water samples, a survey about the main pesticides used at the study areas was performed. The main crops in both sites of study, southeastern and western Buenos Aires, were Wheat and Barley as winter crops; and Soybean, Sunflower and Corn as summer crops. Wheat and Barley use the same herbicides, insecticides and fungicides.

For this study, for economical reasons, twelve of the most common used pesticides for winter and summer crops, and “chemical fallows”, were selected as shown in Table 1. Glyphosate and 2,4-D do not present residual action, while the other nine herbicides present different levels of residual action in soil. Clorpirifos was the only insecticide analyzed for the moment, but cypermetrine and endosulphan are listed for next laboratory analysis.

Table 1-Selected Herbicides for water sample analysis

Active	Type	Main uses
Glyphosate	Herbicide	Soybean, corn, chemical fallows
Iodosulfuron metil	Herbicide	Wheat and Barley
2-4 D	Herbicide	Wheat, Barley, Corn, chemical fallows, etc
Dicamba	Herbicide	Wheat, Barley, Corn, sorghum, chemical fallows, etc
Imazapir	Herbicide	Sunflower and Corn
Triasulfuron	Herbicide	Wheat and Barley
Imazetapir	Herbicide	Soybean
Metilsulfuron-M	Herbicide	Wheat, Barley, Corn, chemical fallows, etc
Atrazine	Herbicide	Corn and sorghum
Fluorocloridona	Herbicide	Sunflower
Acetoclor	Herbicide	Sunflower and Corn
Clorpirifos	Insecticide	Any crop

Water samples were stored at 5 ° C until processing. In the laboratory herbicides study was conducted by solid phase extraction (SPE) using GCB, C18 and PSA (Furlong et al., 2001; Zaugg et al., 1995). The extracts obtained were analyzed with Ultra Performance Liquid Chromatograph (Waters ACQUITY UPLC ®) coupled to a mass spectrometer (Waters ® SQD) (UPLC-MS). Identification was performed with relative retention times to an internal standard and relative abundance ratios of at least two characteristic ions for each analyte. The calibration curves were constructed with solutions of the analytes in solvents prepared gravimetrically with commercial standards.Figures 5 and 6 show the calibrated curves for 2,4-D, dicamba, Imazapyr and Metsulphuron-methil.

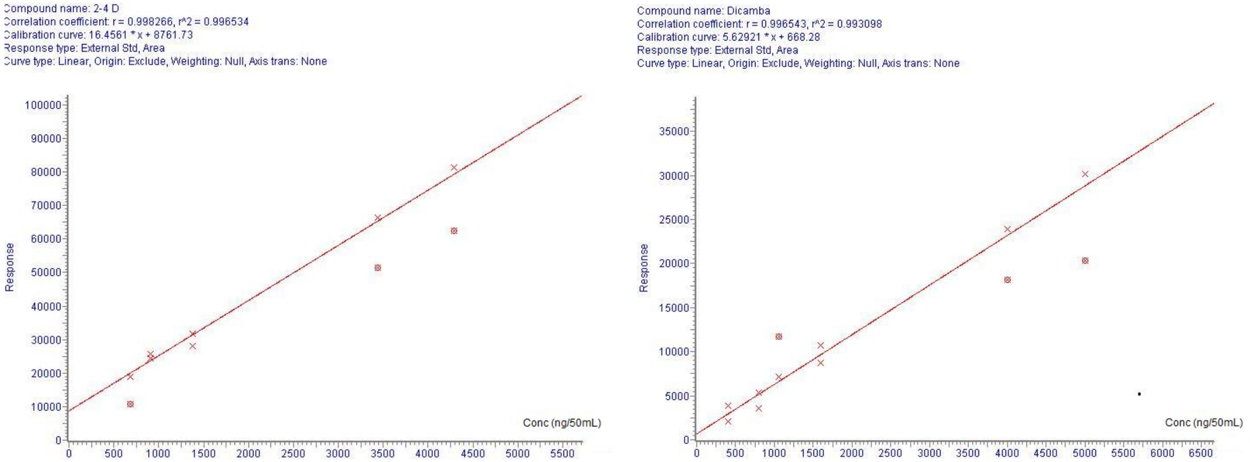

Figure 5 - Calibration curves for 2,4d (Left) and Dicamba (right). Injected mass concentration expressed in nanograms/50 ml

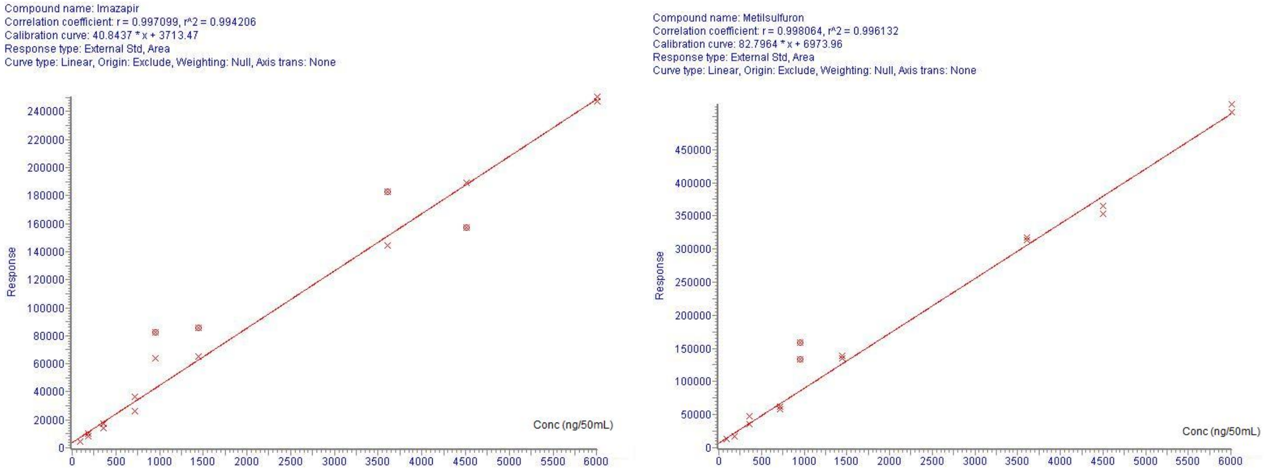

Figure 6 - Calibration curves for Imazapyr (Left) and Metsulphuron-metil (right). Injected mass concentration expressed in Nano-grams/50 ml

For every pesticide the “Limits of detection” and “Limits of quantification” were determined. Limit of Detection (LoD)_ is the minimum amount of analyte that the equipment may detect. Concentrations smaller than LoD cannot be detected. Limit of Quantification (LoQ) is the analyte minimum concentration that can be quantified by the equipment. Table 2 present the LoD and LoQ for the pesticides under study. If a given concentration is higher than LoD and lower than LoQ, the analysis is reported as “< LoQ”, which means that the analyte has been detected, but the amount cannot be reliable measured or quantified.

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Table 2- Limits of detection and quantification for the analyzed chemicals

Analyte	Limit of Detction	Limit of Quantifiaction	Units
Iodosulfuron metil	1.6	4.8	ppb
2-4 D	2.0	6.0	ppb
Dicamba	1.6	4.8	ppb
Imazapir	0.1	0.2	ppb
Thiasulfuron	0.4	1.2	ppb
Imazetapir	0.6	1.9	ppb
Metsulfuron-Methyl	0.6	1.8	ppb
Atrazina	0.2	0.6	ppb
Flurocloridona	0.1	0.4	ppb
Acetoclor	1.0	3.0	ppb
Clorpirifos	0.2	0.6	ppb

Results and Discussion

Groundwater Nitrates concentration

As mentioned above, a second set of 19 observation wells was installed in October 2011. Sampling period extended for 17 months, from November 2011 to March 2013, Table 3 shows the landscape position, crop sequence and total Nitrogen applied, for each observation well.

On average, applied Nitrogen in the season 2011-2012 was higher than applied N in the period 2012-13 because of the present crops at each field. Soybeans did not receive N fertilization and cereals (maize, wheat and barley) were well fertilized with N sources, mainly urea and MAP at planting.

Table 3 -- Site Description for groundwater sampling art western Buenos Aires

Site	Number of wells	Landscape Position	Crop sequence		Total N Applied (KgN /ha)	
			2011-2012	2012-2013	2011-2012	2012-2013
Las Casuarinas	3	Upland- Midslope - Lowland	Maize	Soybean	102	0
Bersee	2	Midslope and Lowland	Soybean	Wheat / Late Soybean	4.4	95
El Estribo	2	Upland and Lowland	Maize	Soybean	82	0
El Porvenir	2	Upland and Lowland	Soybean	Soybean	4	4
El Porvenir	1	Midslope	Soybean	Late Maize	0	66
La Guarida	1	Upland	Sunflower	Maize	64	79
La Guarida	1	Midslope	Maize	Soybean	93	0
La Guarida	1	Lowland	Maize	flooded	93	0
Los Alamos	2	Upland andMidslope	Barley/Late Soybean	Soybean	86	0
Los Alamos	1	Lowland	Barley/Late Soybean	flooded	86	0
San Jorge	1	Upland	Barley/ Late Corn	Soybean	104	0
San Jorge	1	Midslope	Barley	Soybean	72	0
San Jorge	1	Lowland	Sunflower	Barley	6	113
Total Wells	19		Average Applied N (kg/ha)		61.3	27.5

Therefore, considering a higher N input in the first crop season (2011-2012), a higher N loads would be expected in groundwater during the first summer. However, the higher concentration at groundwater was recorded in winter and spring 2012, because of the occurrence of unusual high rainfalls, causing flooding at many farm fields from August to November 2012.

Rainfall events from May to October 2012 added 601 mm, and the mean precipitation for that period between 1953 and 2010 was 279 mm. This amount of precipitation fell during winter and early spring, caused that groundwater table reached the soil surface causing flooding in October and November, increasing nitrate leaching significantly starting in May (Figures 7 and 8).

As also expected, for sandy soils, rainfall amount and frequency was the main driver for N transport to phreatic aquifer, rather than N fertilization rates, landscape position o groundwater table proximity (Figure 10).

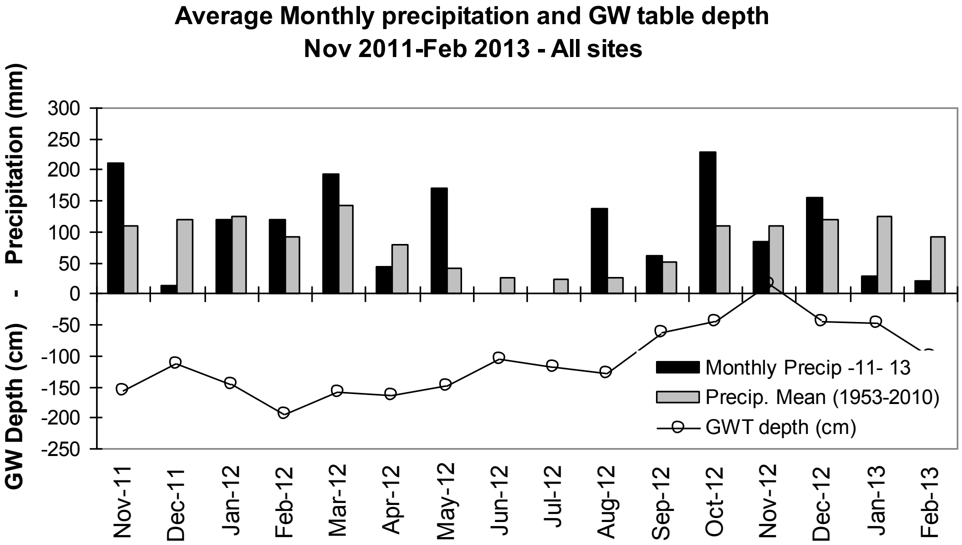

Figure 7 - Historical rainfall records and monthly precipitation and GW table depth for the study period

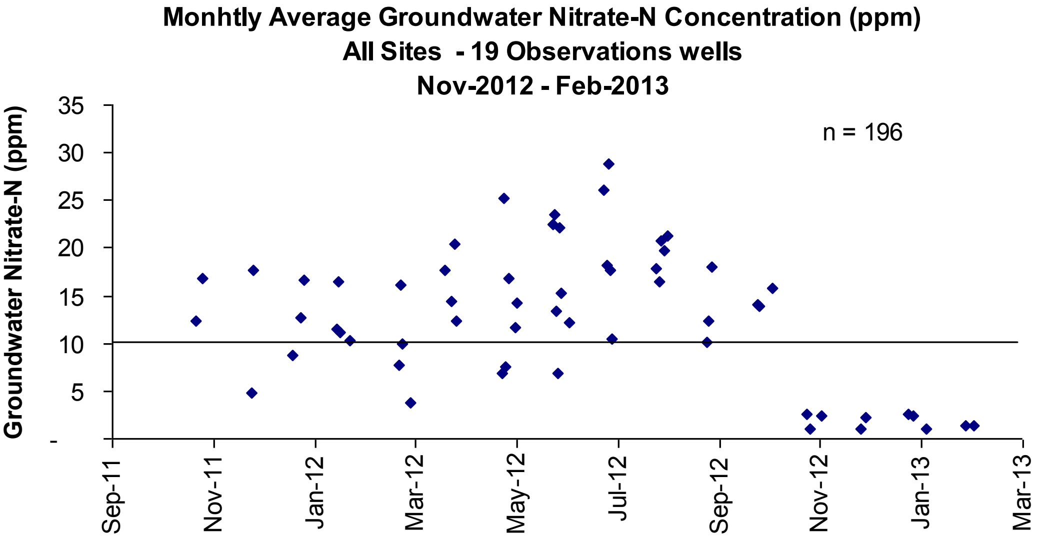

Figure 8 – Monthly average Groundwater nitrate-N concentration for the study period for all sites (n=196). Horinzontal line at 10 ppm, indicates EPA critical for drinking water

There were significant differences in nitrate concentration and water table depth, due to landscape positions. Nitrate-Nitrogen concentrations were higher in upland wells than in midslope and lowland wells. Figure 6 shows separately the average monthly groundwater table and NO_3-N concentration for all the three landscape positions. Because of high precipitations, both groundwater table depth and nitrate leaching increased until November, and then they normalized during summer, presenting very low values of NO_3-N, around 1.5 mg/L.

Differences between NO_3-N at lowland and upland positions, may be explained by effect of water dilution. Nevertheless, some other processes such as mineralization and denitrification, also related to soil N dynamics, have to be considered, especially because NO_3-N levels were not related to N fertilization rates.

High values found at upland wells and low values at lowlands, as well as an average increasing in Nitrate concentration might be also due to natural processes of mineralization and denitrification, along with nitrate leaching to groundwater. Even in absence of agriculture, mineralization from soil organic matter increases nitrate soil levels. This process depends on bacterial activity which directly relates to temperature and soil moisture. On the other hand, denitrification reduces nitrates to nitrous oxide (N_2O) and nitrogen gas (N_2) in soil saturation conditions. In this way, in Pampa region soils, Portella et al.(2006) used N^{15} to keep track of leached N coming from fertilizer at corn fields in two texture soils: clay loam and sandy loam. Even though leached N was very low and similar for both soils, the authors reported that there was a low contribution of fertilizer N (0–3.5%), implying that >96% of the leached N was derived from soil organic matter mineralization.

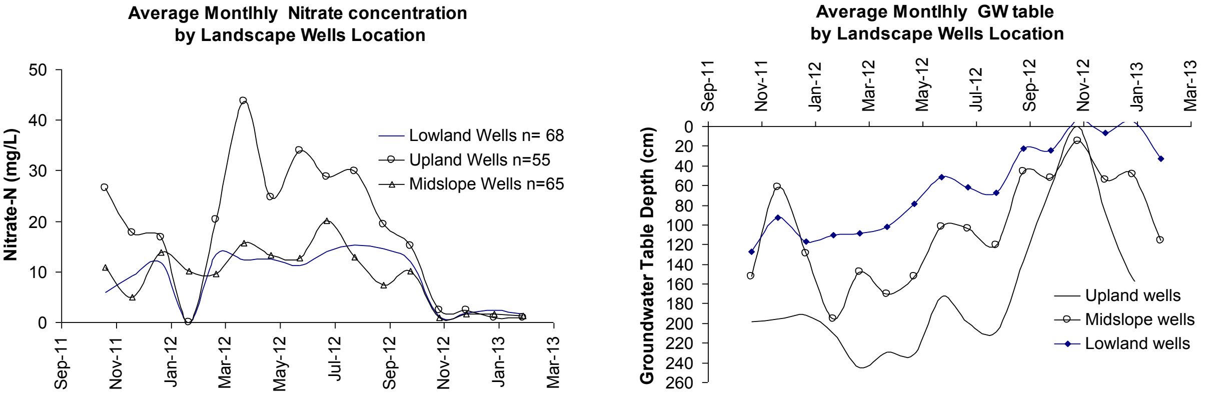

Figure 9 - Average monthly NO3-N concentration and Water Table depth by landscape position.

<u>Surface Water Nitrate concentration</u>

Preliminary results for this variable, showed an average nitrate-Nitrogen concentration in streamflow between 4 and 5 ppm. Samples from Napaleofu and Quequen Grande streams, presented similar average values. Figure 10 depicts a time series for NO_3-N concentration at Quequen River from September to November 2011. For that period, NO_3-N concentration was always below the critical value for drinking water of 10 ppm.

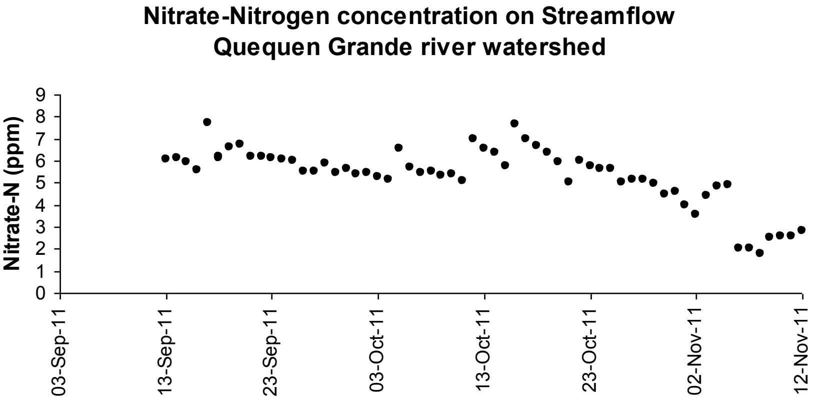

Figure 10 - Nitrate-N concentration on streamflow for Quequen Grande River watershed (Septembre 2011- November 2011)

Groundwater and Surface Water Chemical Residues

A big number of samples are under analysis; however a subset of samples was separated to test the equipment and the calibration curves. Therefore, preliminary results are presented only for a subset of 100 samples that were analyzed. A first subset of 48 samples belongs to surface water: 24 samples from Quequen Grande River and 24 from Napaleofu Creek. All of them represent a bi-weekly sample frequency for the period October 2011-September 2012.

A second set of 52 samples was selected out of 190 monthly groundwater samples collected from November 2011-April 2013.

As for surface water samples, most of theml were negative (no detection) for any pesticide. Only 5 out of 50 samples presented small concentrations of clorpirifos and acetoclor. Table 4 shows the results and sampling date of the mentioned samples.

Table 4 - Detail of the results of the five samples from streams which presented any chemical in solution

Watershed	Chemical detected	Concentration (ppb)	LoD (ppb)	LoQ (ppb)	Sampling Date
Napaleofu	Clorpirifos	< LoQ	0.2	0.6	30-Oct-11
Napaleofu	Clorpirifos	< LoQ	0.2	0.6	22-Feb-12
Napaleofu	Clorpirifos	4.3	0.2	0.6	12-Sep-12
Quequen	Acetoclor	< LoQ	1	3	30-Apr-12
Quequen	Clorpirifos	< LoQ	0.2	0.6	10-Aug-12

Regarding to groundwater, atrazine and clorpirifos were detected in 11 samples, out of 50 analyzed samples, but 10 of them in concentrations less than the limit of quantification (< LoQ). In only one groundwater sample clorpirifos showed a quantifiable value at 3.3 ppb (Table 5). Even when these results are very preliminary, chemicals residues detected at groundwater and streams were almost negligible and most herbicides used where not even detected as traces. However, clorpirifos, an insecticide, was the most frequently detected along with atrazine. Present results are useful to start monitoring water bodies in order to protect water sources. Best management practices are a key issue for preventing nutrient and chemical losses to water bodies.

Table 5 – Detail of chemicals detected in groundwater samples

Sample #	Observarion Well	Chemical Detected	Concentration (ppb)	LoD (ppb)	LoQ (ppb)	Sampling Date	Present crop
1	F16	Atrazine	< LoQ	0.2	0.6	6-Jan-12	Soybean
	F16	Clorpirifos	< LoQ	0.2	0.6	6-Jan-12	Soybean
2	F11	Clorpirifos	< LoQ	0.2	0.6	4-Jan-12	Corn
3	F14	Clorpirifos	3.3	0.2	0.6	4-Jan-12	Corn
4	F11	Clorpirifos	< LoQ	0.2	0.6	25-Jan-12	Corn
5	F1	Atrazine	< LoQ	0.2	0.6	26-Jan-12	Barley
6	F17	Atrazine	< LoQ	0.2	0.6	3-Nov-11	Soybean
7	F13	Atrazine	< LoQ	0.2	0.6	6-Dec-11	Corn
	F13	Clorpirifos	< LoQ	0.2	0.6	6-Dec-11	Corn
8	F8	Clorpirifos	< LoQ	0.2	0.6	8-Dec-11	Soybean
9	F19	Atrazine	< LoQ	0.2	0.6	6-Nov-11	Sunflower
10	F18	Clorpirifos	< LoQ	0.2	0.6	6-Nov-11	Corn
	F18	Atrazine	< LoQ	0.2	0.6	6-Nov-11	Corn
11	F20	Acetoclor	< LoQ	1	3	6-Nov-11	Corn
	F20	Clorpirifos	< LoQ	0.2	0.6	6-Nov-11	Corn

NPS Pollution Model Validations: advances in hydrological components.

SWAT model was chosen as watershed scale model that includes subroutines for pesticide and nutrient transport and other useful subroutine for agricultural watersheds**.**

The SWAT (Soil Water Assessment Tool) model (Arnold et al., 1998) allows simulation of the impact of different scenarios on the levels of atrazine over time and space. Thus, SWAT constitutes a valuable tool to study the impact of fertilizer and pesticide use on water sources, as well as the impact of management practices and potential land use changes. In previous evaluations, SWAT has shown good results when predicting runoff (Saleh et al., 2000; Spruill et al., 2000) and nitrogen and phosphorus levels in streams (Saleh et al., 2000; Saleh and Du, 2004). SWAT daily predictions for atrazine were evaluated in Sugar Creek, Indiana (242 km2) by Neitsch et al. (2002), who reported a daily R2 of 0.21 and 0.41 in the calibration and validation periods, respectively. At the St Joseph River Watershed, SWAT closely predicted, the total mass of atrazine released by the whole basin between 2000 and 2003, for the period April to September (Vazquez-Amabile et al., 2006).

In southeastern Buenos Aires, SWAT model has been recently validated for streamflow at Quequen Grande river watershed, as a part of an international bigger project focused on the evaluation of changes in water productivity against potential future scenarios of climate change.

Model predictions for monthly and daily streamflow are presented in table 6. Previous to start using the model for nutrient and pesticide calibration, daily calibration for streamflow is very important. Daily predictions for this watershed were acceptable, since there was not a representative network of rain gages to cover the almost one million hectares of the watershed. Monthly predictions are presented in Figure 11 for both calibration and validation periods.

Table 6 - Results for daily and monthly streamflow SWAT model calibration and validation, at Quequen Grand River watershed

	Calibration 1996-2000		**Validation 2001-2006**	
Período	Daily	Monthly	Daily	Monthly
Observed Streamflow (m^3/s)	16.57	16.54	40.42	36.01
SWAT Streamflow (m^3/s)	16.67	16.7	37.11	36.58
$\mathbf{R^2_{Nash}}$	0.21	0.75	0.37	0.61
Pearson coef. of Correlation	0.48	0.72	0.64	0.8

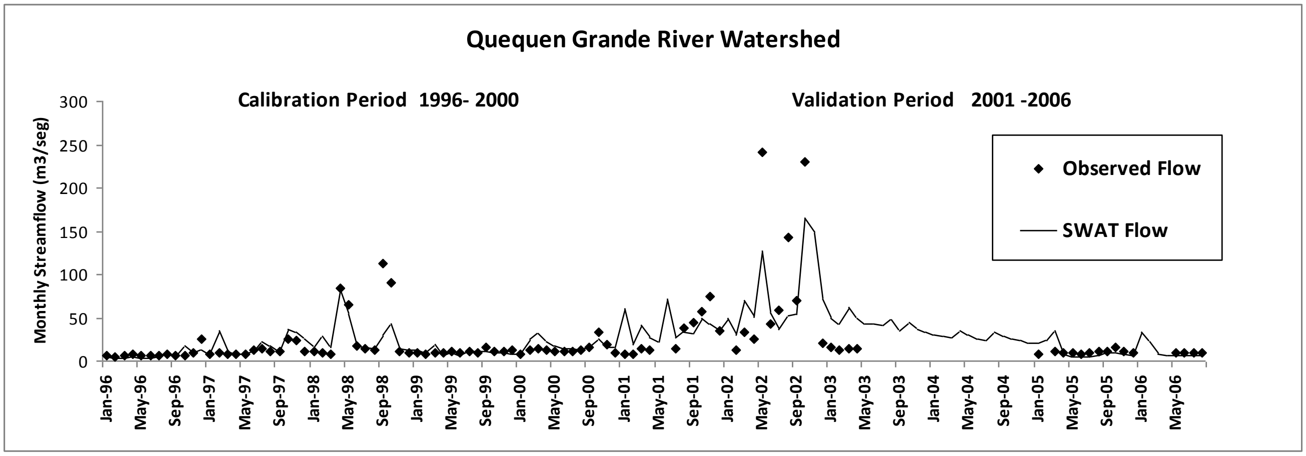

Figure 11 - Time series for Observed and Predicted Monthly streamflow, at Quequen Grande River Watershed, for CAlibratin and Validation periods

Summary and conclusions

Non-point source water pollution is a key issue in rural watersheds and it needs to be studied in order to prevent damages to ground and surface water quality. This is especially important in Argentina where the use of chemicals in agriculture has increased in the last years, due to the increasing adoption of No Tillage system and the expansion of agricultural crops.

Preliminary results were presented for two sites of study , in order to show a first picture about subsurface and streamflow water quality. They are presented from a subset of samples, since remaining samples are currently being processed in laboratory.

Regarding to NO_3-N concentration, most of wells presented variable concentration depending on monthly precipitation and landscape position. Considering 10 mg/L NO_3-N as a standard limit, 52% of the observations exceed this value mostly related to unusual precipitations events at winter 2012. Thus, results showed that rainfall amount was the main driver for N transport to phreatic aquifer, rather than N fertilization rates, landscape position o groundwater table proximity

Analyzed samples from surface water showed an average Nitrate-N concentration in streamflow of approximate 5 ppm at Quequen Grande River and Napaleofu creek. .

A second goal of this on-going research is the validation of the GLEAMS and the SWAT hydrologic models. Both models might be a valuable tool for studying risk of NPS pollution under different soils, management practices and climate scenarios in agricultural lands of Pampa region, Argentina. In this sense, some results about the validation of the hydrological component of SWAT were presented for Quequen Grande River Watershed. This model might be a useful tool for researchers and decision makers to study the impact of potential future changes such as land use or climate change.

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