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Article

Spatial and temporal distribution pattern of phosphorus fractions in a saline lowland river with agricultural land use (Salado River, Buenos Aires, Argentina)

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With 7 figures and 6 tables

Abstract: The main river of the Buenos Aires province, the Salado (length, 600 km; basin area, 150,000 km^2), which drains one of the country's most extensive agricultural regions, is impacted by human activities (agriculture, hydraulic modifications), although the extent and mechanism of those impacts have yet to be clarified fully. This study investigated the temporal and spatial variations in concentrations of different phosphorus fractions in the Salado and its channels and tributaries to examine the potential influences of land use on the phosphorus dynamics in the basin. The data collected between 1997 and 1999 – during which period the basin experienced El Niño and La Niña events – indicated that the main phosphorus input occurred upstream and was mainly attributable to agricultural runoff. In support of this notion, the timing of the spring peak of total phosphorus concentrations (3,500 µg L^{-1}) corresponded to the period of highest agricultural activities in a tributary of the headwaters. Of the total dissolved phosphorus, the particulate phosphorus and the dissolved reactive phosphorus were the main fractions in the headwaters. Data were also collected in the lower sector of the basin during the flooding periods between 2002 and 2003. These results revealed that a large total-phosphorus transport in the spring was related to a high discharge as a result of heavy rainfall. As with the results obtained in the entire basin, the particulate phosphorus fraction was primarily responsible for the increase in total phosphorus. We furthermore found that the phosphorus concentrations decreased near the mouth of the Salado, where extensive wetlands were present.

Key words: phosphorus fractions, lowland river, agricultural land use, land use index, Pampa region, Argentina.

Introduction

Rivers and streams are the major routes of phosphorus (P) transfer to lakes and oceans. These lotic bodies unite the heterogeneous losses of P from their catchments, while biogeochemical and physical processes within the moving water modify the forms of P. The transport rate of P in streams and rivers depends on the forms and concentrations of P and the discharges (Melack 1995). Excessive inputs of P could result in eutrophication, causing problems for domestic-, industrial-, and recreational-water use as well as for the ecological status of the region. Resolving these problems represents one of the key challenges for attaining environmental sustainability.

In general, rural land use can be a major nonpoint source of P and sediment for both lotic and lentic waters. Extensive studies have been conducted to exam-

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ine the relationship between P inputs, agricultural land use and soil-related processes (Carpenter et al. 1998, Haygarth et al. 2005). In Argentina, however, knowledge is limited concerning the relationship between changes in land use and P dynamics within major river basins. A comprehensive monitoring of P over space and time is needed in order to examine variations in the concentrations of the different forms of P under variable hydrological conditions.

Currently in Argentina, 80 % of the fertilizers are composed of urea, ammonium, and diammonium phosphates. The continuous increase in P application between the 1960s and the 1980s came mainly from the autochthonous soil P. During the first years of the 1990s, the use of P per ha was between 10 to 15 kg. Soybean crop was the main P source producing this increase (Stauffer & Sulewski 2003). In the pampean region, the soils have always had P in their composition (Morras 1999); but with the increases in agricultural activities, the fertilizers have become more influential in the transfer of P into the water bodies. Although the fertilization served as an aid in increasing the yield per ha, studies on the impact of this agricultural practice on the aquatic environments of the region have not been undertaken. The data on P dynamics are generally scarce in developing countries, even though such data are required for establishing control and management policies of nutrient salts (Withers & Haygarth 2007).

Climatic variability may largely affect agricultural production in Argentina. The pampas is characterized by high variability in rainfall from year to year. The El-Niño-Southern-Oscillation (ENSO) phenomenon accounts for much of this fluctuation in meteorological conditions (Messina et al. 1999). The variation in maize yields in Argentina is closely related to the ENSO event, with December and January being influential months for determining the final production from the harveSts Soybeans exhibit a significant association with the ENSO cold phase (although the frequency of the cold event is low within a typical fluctuation cycle). A clear association exists between negative-precipitation anomalies and soybean yields. Sunflower yields do not evince an apparent association with the ENSO phases (Podestá et al. 1999). When the strong 1997 El Niño event resulted in enhanced precipitation throughout the pampas, yields for maize and soybeans (after accounting for trends in technology) were high (Podestá et al. 1999). According to Bettolli et al. (2009), soybean yields are related negatively to La Niña events.

Human interventions have affected the integrity of regional ecosystems in the pampas during the last century (Viglizzo et al. 2001). The region has a relatively short farming history since the pampas had remained as native grassland until the end of the 19th and the beginning of the 20th centuries. Cattle production was a predominant activity during most of the 20th century, but crop production continually increased from the 1960s on (Viglizzo et al. 1997). Recent results (Viglizzo et al. 2004) demonstrate that the ecological functions had been entirely controlled by natural environmental forces during the postcolonial period (before 1880), when human colonization and the corresponding ranching process had not yet begun. Once both processes were triggered and a typically extensive, low-input farming model was consolidated between 1880 and 1960, anthropogenic forces increased in their impact and became scaled up, counteracting in part the natural controlling effects of the environment. Such human-driven control increased quickly during the period of greater intensification (1960–2000). The spatial aggregation of such anthropogenic effects in plots and farms in the 1990s appeared to be strong enough to minimize the influence of natural controls over large geographical areas (Viglizzo et al. 2004).

The spatial distribution of the various P-input sources over a given landscape creates a complex and dynamic mosaic of potential P inroads, especially since those inputs vary within the agricultural year (e.g., the timing of the manure and fertilizer applications (Haygarth et al. 2005)). In contrast to point P sources, diffuse P losses from agriculture are spatially and temporally highly variable and thus difficult to quantify and trace back to specific fields. This P originates from a number of different areas within the catchment, is transported by diverse hydrologic pathways, and arrives at the watercourse at varying times, often some distance from the source when biologic P-fixing activity is reduced (Hodgkinson & Withers 2007). In recent years, nonpoint sources of P are the dominant inputs into most of the surface water of the USA and are mainly related to the presence of croplands (Carpenter et al. 1998). The contribution of fertilizer runoff varies from near 0 to 100 % of the input among river systems according to Caraco (1995).

In the Salado-River basin, the land use shows a clear gradient, being more intensive in the headwaters with respect to agricultural activities (mainly soybean, sunflower, maize, and wheat; Gabellone et al 2005). The basin is characterized by high concentrations of P and the presence of phytoplankton species that indicate eutrophication (Neschuk et al. 2002). The total P concentration bears a direct relationship to the distance from the mouth (Gabellone et al. 2005).

Detailed spatial and temporal analyses of different P fractions help examine relationships between land use and the quantity and quality of P in rivers. Knowledge of the forms of P is essential for establishing a basis for the reduction and control of P sources (Donohue et al. 2005). The aim of this study was to determine the relationship between the spatio-temporal dynamics of the different P fractions in the main channel and tributaries of the Salado River and the land use during the years 1997–1999, coincident with major changes in ENSO events, which could constitute an important driver of watershed runoff in the region. An exceptional flooding period occurring three years later (2002–2003) was likewise investigated to analyze the fractions within the P input entering the Salado mouth from the lower sector of the basin at that time.

Material and methods

Study area

The Salado River is the southernmost tributary of the Río de la Plata basin and the major river arising within the Buenos Aires province (Conzonno et al. 2001) with a length of 600 km and a basin area of 150,000 km^2. The more humid conditions in the second half of the last century promoted the formation of extensive wetlands following a definite pattern within the pampean plains (Iriondo 2004). The Salado River basin is located in a dry, temperate flatland (Gustard 1994) and includes

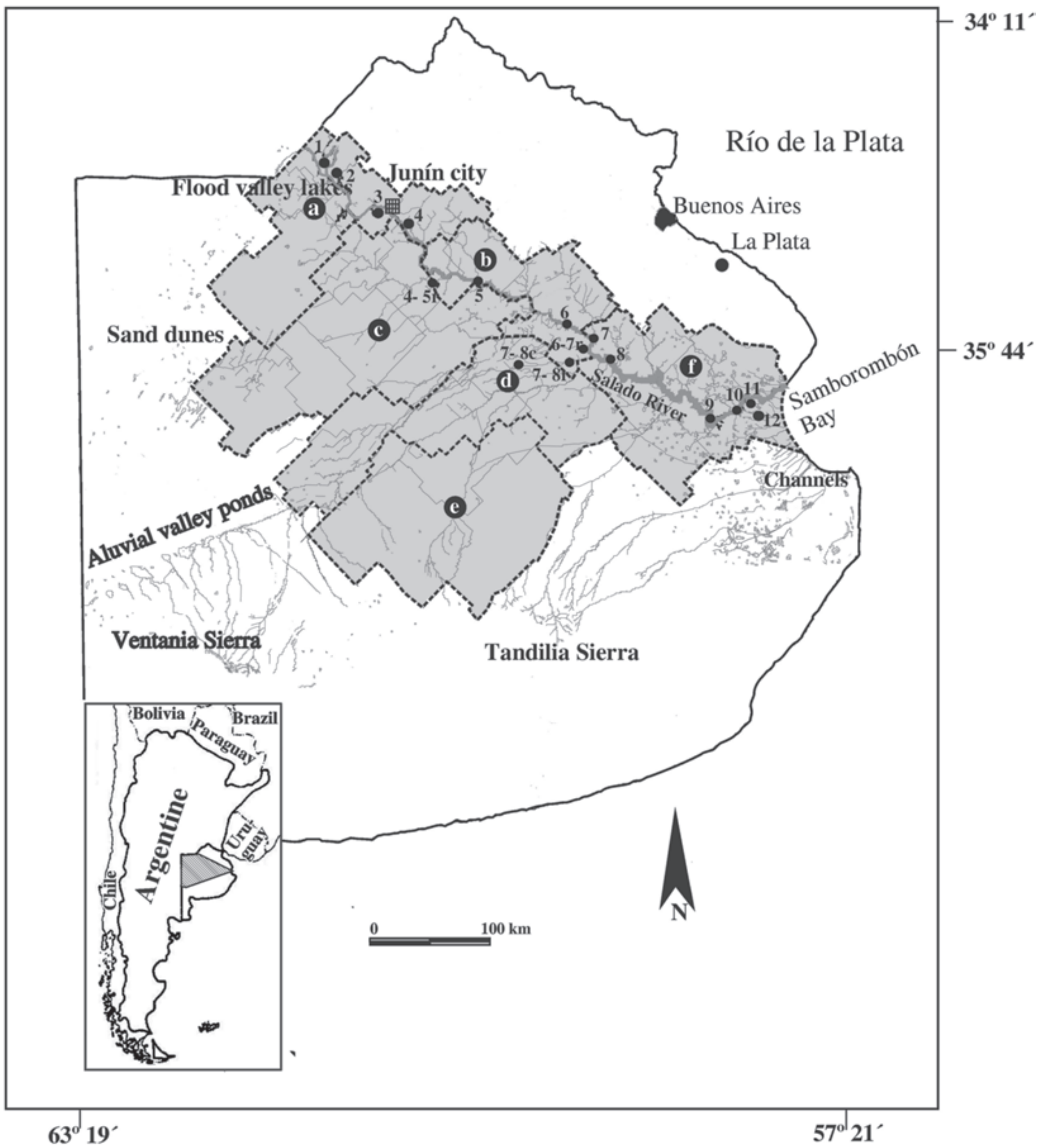

Fig. 1. Map of the Salado River basin showing the land-use zones (lower-case letters) and the sampling stations (numbers).

a large number of shallow lakes occupying 10,000 km^2 under normal conditions of river flow. The regime of the Salado River is quite variable from a flow reaching no more than 100 m^3 s^{-1} in dry periods to as much as 1,500 m^3 s^{-1} during floodings, with corresponding variations in conductivity and the transport of dissolved and particulate materials. The flooding of large areas during weeks or months is one of the most salient characteristics of the pampean plains.

The Salado River has a gradient in the intensity of the land use: intensive agriculture in the headwaters and extensive livestock in the lower basin. The river has very little slope and its bottom is soft (clay and silt). The population density is low, and the urban and rural populations do not have waste water service and use septic tanks. In cities such as Junín, located at headwaters with 64,000 inhabitants, the effluent of the sewage treatment (secondary) discharges in lentic environments. In general, the river has natural buffer zones that correspond to flooding areas. However, in the headwaters, these areas may be impacted by agricultural activities (for more information about geological features and land use: Gabellone et al. 2005, Gabellone et al. 2008).

Field sampling

Regional study

In the northern sector of the basin, five sampling stations were established: Station 1 (St. 1) at the Salado stream; Station 2 (St. 2) at the Piñeiro stream; Station 3 (St. 3) at Junín1; Station 4 (St. 4) at Junín2; station 4-5r (St. 4-5r) at the Saladillo stream where it receives the Jauretche-Mercante República de Italia canals; and Station 5 (St. 5) at Achupallas. In the western middle sector of the basin, three sampling stations were also established: Station 6 (St. 6) on Ruta 30; station 6-7r (St. 6-7r) at the Saladillo–Vallimanca stream (in a subbasin), one of the main tributaries of the Salado River and originating in the Ventania sierras; and Station 7 (St. 7) at Roque Pérez. In the region referred to as the Depressed Pampa, seven sampling stations were likewise established: station 7-8r (St. 7-8r) at the Las Flores stream (subbasin), whose source lies in the Tandilia sierras; Station 7-8c (St. 7-8c) at Canal 16, which body regulates the discharge of the Vallimanca stream; Station 8 (St. 8) on the Gorchs' left bank; Station 8 r (St. 8 r) on the Gorchs' right bank; Station 9 (St. 9) at General Belgrano; Station 10 (St. 10) at El Destino; and Station 11 (St. 11) at Guerrero, located 84 km from the mouth of the Salado River. No sampling stations were established downstream from Station 11 because below that point 80 % of the river discharge runs through two canals, starting at 69 km from the river mouth (Canal 15 and Canal Aliviador). The tributaries are named with the numbers of the stations of the main course located upstream and downstream of each affluent. The letter "r" in the tributaries indicates that the stream is an affluent of the right bank of the river. The letter "c" refers to the artificial condition of the tributary (i.e., a canal; Fig. 1).

The sites were visited initially in March 1997, and then quarterly on a seasonal basis through the autumn of 1999. On the first four occasions, 14 stations were sampled. During the period of May 1998–June 1999, two further tributaries were

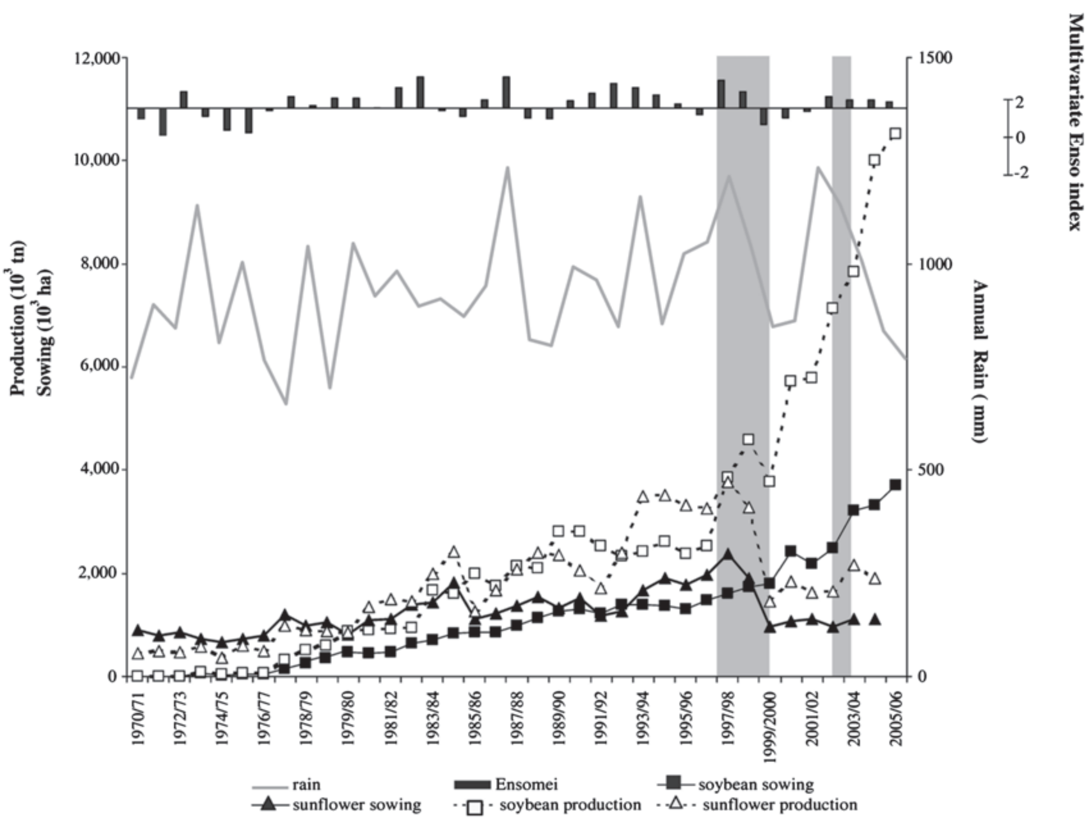

Fig. 2. Historical data on the series of multivariate ENSO index (MEI) events, sunflower and soybean sowing area and harvest data, and rainfalls of the Buenos Aires province. The gray bars indicate the sampling periods.

Table 1. Characterization of the zones defined within the Salado River basin, according to the subbasin, with respect to population (mean, and range in brackets), land use data, the calculated Land Use Index and the representative sampling stations.

Zone	Population density	Number of departments	Total area	Crops area	Cattle area	Land Use Index	Sampling stations
	(inhabitants km^{-2})		(km^2)	(%)	(%)	(LUI)	
a	12.1 (7.1–39.4)	5	13,408	53.0	47.0	1.13	1, 2, 3, 4
b	14.9 (6.9–29.3)	5	8,114	29.2	70.8	0.41	6, 7
c	10.2 (7.3–18.2)	6	15,931	34.5	65.5	0.53	4-5r, 5
d	7.0 (3.2–11.0)	4	14,321	7.9	92.1	0.09	6-7r, 7-8c
e	8.4 (1.97–13.5)	3	18,348	12.6	87.4	0.14	7-8r, 8r
f	7.9 (3.8–9.3)	5	11,114	4.4	95.6	0.05	8, 9, 10,11

Site 12 is not incorporated in Table 1 because it was not sampled in the LU analysis, and the information obtained there was only used in the flood condition analysis.

also included because they bring in water from the subbasins of the surrounding sierras (Sts 7-8c and 7-8r; Fig. 1). The inclusion of these sites improved the interpretation of the results. The sampling period was characterized by the highest principal El-Niño event (1997–1998, Fig. 2) followed a La-Niña occurrence (1998–1999). Nevertheless, this global weather pattern failed to raise the river's discharge above the average values. The Multivariate El-Niño–Southern-Oscillation Index (MEI; Wolter & Timlin 1993) was used for the comparison between rainfalls and soybean or sunflower yields. The values of ENSO MEI were obtained in www.esrl.noaa.gov/psd/enso.

The data on discharge are scarce both in terms of the frequency and the location of the measurements. Only four gauges are available with a few further data garnered from the to Dirección de Hidráulica del Ministerio de Obras Públicas de la Provincia de Buenos Aires.

Flooding conditions in the lower-basin sector

Four samplings were performed each season during the period from May 2002 through February 2003 for assessing the effects of flooding on P transport. The flow discharge achieved during that period was five times the mean value; with a recurrence, related to exceptional rains in the basin – those occurring historically at an interval of every 10 years in coincidence with a La-Niña event (Fig. 2). Four sites located at the lower sector of the river were monitored on two consecutive days. Out of the entire study, four stations were included: Station 9, 35° 43′ S, 58° 31′ W, General Belgrano; Station 10, 35° 57′ S, 58° 00′ W El Destino; Station 11, 35° 57′ S, 57° 51′ W, Guerrero; and Station 12, Canal 15, 35° 58′ S, 57° 27′ W.

The Dirección de Hidráulica del Ministerio de Obras Públicas provided the discharge data for the various sampling occasions. The rain data for locations nearest the sampling sectors were obtained from databases of the Secretaría de Agricultura, Ganadería y Pesca de la Provincia de Buenos Aires (SAGYP): Belgrano city (35° 46′ S, 58° 29′ W); Pila Village (36° 00′ S, 58° 08′ W); and Castelli village (36° 05′ S, 37° 48′ W). In addition, precipitation data for the regions representative of the subbasins were collected: Junín city (34° 34′ S, 60° 57′ W) and Azul city (36° 46′ S, 59° 51′ W). Certain localities – Junín, Azul, General Belgrano, and Dolores (36° 18′, 57° 40′ W9) – that had normally experienced rainfall daily in the past (1911–2002) were used to compare each three-month mean value with the figure from that corresponding sampling period.

Zones of land use

Six zones (Fig. 1) of Land Use (LU) were defined in the Salado River according to their subbasin characteristics (PRS 1999). Each region included a different number of administrative areas (i.e., departments). The population density, the total area, and the cattle and crop production of every department were obtained from the Secretaría de Agricultura, Ganadería y Pesca de la Provincia de Buenos Aires. The crops included were soybean, wheat, sunflower, and maize. The cattle production ranged from cattle-breeding to steer-fattening and dairy operations. The data are expressed as the average value over the 6-year period from 2000 through 2005. The corresponding data from each zone represent the summation of the crop and cattle areas as a percent of the total area of the departments included. The land-use index (LUI) was calculated as the ratio between the total crop area and the cattle-production area, expressed as percents of the total area. The tributaries, representing subbasins, were included in each zone according to their headwater locations: for example the sampling Stations 7-8 and 7-8r correspond to Zone E. Table 1 summarizes those data for each zone.

Laboratory procedures

At each sampling, the water comprising the first 0.5 m of the water column from the surface was taken from midstream and collected in 1-L acid-cleaned polyethylene bottles, transported back to the laboratory in an ice-cooled isolation box, and stored in the dark at 5–8 °C before analysis. Before transportation, all samples were passed through Whatman GF/C filters to increase the subsequent filtration rate because the samples were to be employed for estimations of P fractions, chlorophyll-*a*, and ionic composition (Gabellone et al. 2008). Once in the laboratory, the subsamples for P determination were filtered through Whatman GF/F filters. The subsamples were analyzed for total reactive P (TRP), dissolved reactive P (DRP), total dissolved P (TDP) and total P (TP). To determine TRP and DRP water samples were assayed colorimetrically according to APHA (1995).

Table 2. Methods for phosphorus fractions analysis and the nomenclature employed in the text.

	Name	Method
Without filtration	Total Phosphorus (TP)	1. Persulfate Digestion 2. Colorimetry (APHA 1995)
	Total Reactive Phosphorus (TRP)	Direct colorimetry (APHA 1995)
Filtrate	Total Dissolved Phosphorus (TDP)	1. Persulfate digestion 2. Colorimetry (APHA 1995)
	Dissolved Reactive Phosphorus (DRP)	Direct colorimetry (APHA 1995)
Calculated	Particulate Phosphorus (PP)	TP – TDP (APHA 1995)
	Particulate Organic Phosphorus (POP)	(TP – TDP) – (TRP – DRP) (APHA 1995)
	Soluble Unreactive Phosphorus (SUP)	TDP – DRP (House & Denison 1998)
	Particulate Reactive Phosphorus (PRP)	TRP – DRP (APHA 1995)
	Particulate Unreactive Phosphorus (PNRP)	TP – TRP (Svendsen et al. 1995)
	P in Suspended material (PSS)	PP/SS (in µg P mg^{-1}; House & Denison 1998)

TDP and TP were converted to DRP by acid-persulfate digestion and TRP and DRP determined according to APHA (1995). The concentration of suspended solids (SS, in g L^{-1}) were measured according to method 2540 D of APHA (1995). The concentrations of particulate organic matter (OM) were determined by weight loss after ignition at 550 °C (method 2540E, APHA 1995). The concentration of P in the suspended material (PSS) is particulate P (PP)/SS (in µg P mg^{-1}; House & Denison 1998). Table 2 summarizes the determination and calculation of the various P fractions.

Data analysis

In the regional sampling, to analyze the relationship between distance of the river to the mouth and the P fractions present, we performed a product-moment correlation. Multiple-regression analyses (stepwise method) was effected to analyze the relationship between the land-use index units (LUIs) and the P fractions, OM, and SS. The data used corresponded to the mean values for the sampling stations included in each LUI through the entire sampling period and according to season. The K-means–clustering technique (Dufrêne & Legendre 1997) was used for grouping the sampling stations according to the mean concentrations of four P fractions (TP, TDP, DRP, PP) and PM. Cluster analysis was performed through the use of a standardized matrix of mean values per station for all the data on the fractions of P, OM, and SS as well as the distance of each sampling station from the river mouth. The complete-linkage method and the Euclidian distance were used.

A one-way ANOVA was performed for determining the possible differences in TP concentrations between the consecutive samplings carried out during the flooding period (2002–2003).

The daily TP load was calculated by multiplying the measured concentrations with the instantaneous river discharge at the sampling point.

Results

Regional sampling

Mean and range of the concentrations of different P fractions are tabulated in Table 3. The maximum concentrations of TP were detected at headwater (LU a) stations including St. 4-5r (ISp; hereafter, the sampling occasion when the observation was made is indicated in parenthesis using the identification code as explained in the legend of Fig. 3a) and St. 4 (IS), while the minima were recorded in the tributary (LU d and e) stations including Sts 7-8r and 7-8c (IA) (Table 3). The maximum concentrations of TDP were observed at St. 4-5r (ISp) and St. 4 (IS). The lowest values of TDP concentrations were found at stations located in the tributaries (Sts 7-8r and 7-8c). The DRP maxima were recorded at St. 4 (IS) and St. 4-5r (ISp), while the minima were found at St. 8 (ISp) and St. 7-8r (IA). The highest and lowest values of PP were found at St. 3 (W) and St. 4-5r (ISp), respectively. At stations receiving water from the tributaries of LU d and e, PP concentrations were occasionally (typically in spring and autumn) very low (< 30 µg L^{-1}). The highest concentration of PRP was found at St. 4 (W) and St. 4-5r (ISp). In the summer of 1998, PRP concentration was below the detection limit at six stations (data not shown). The maximum levels of soluble unreactive P (SUP) were observed at Sts 4 and 4-5r (ISp). The minimum SUP

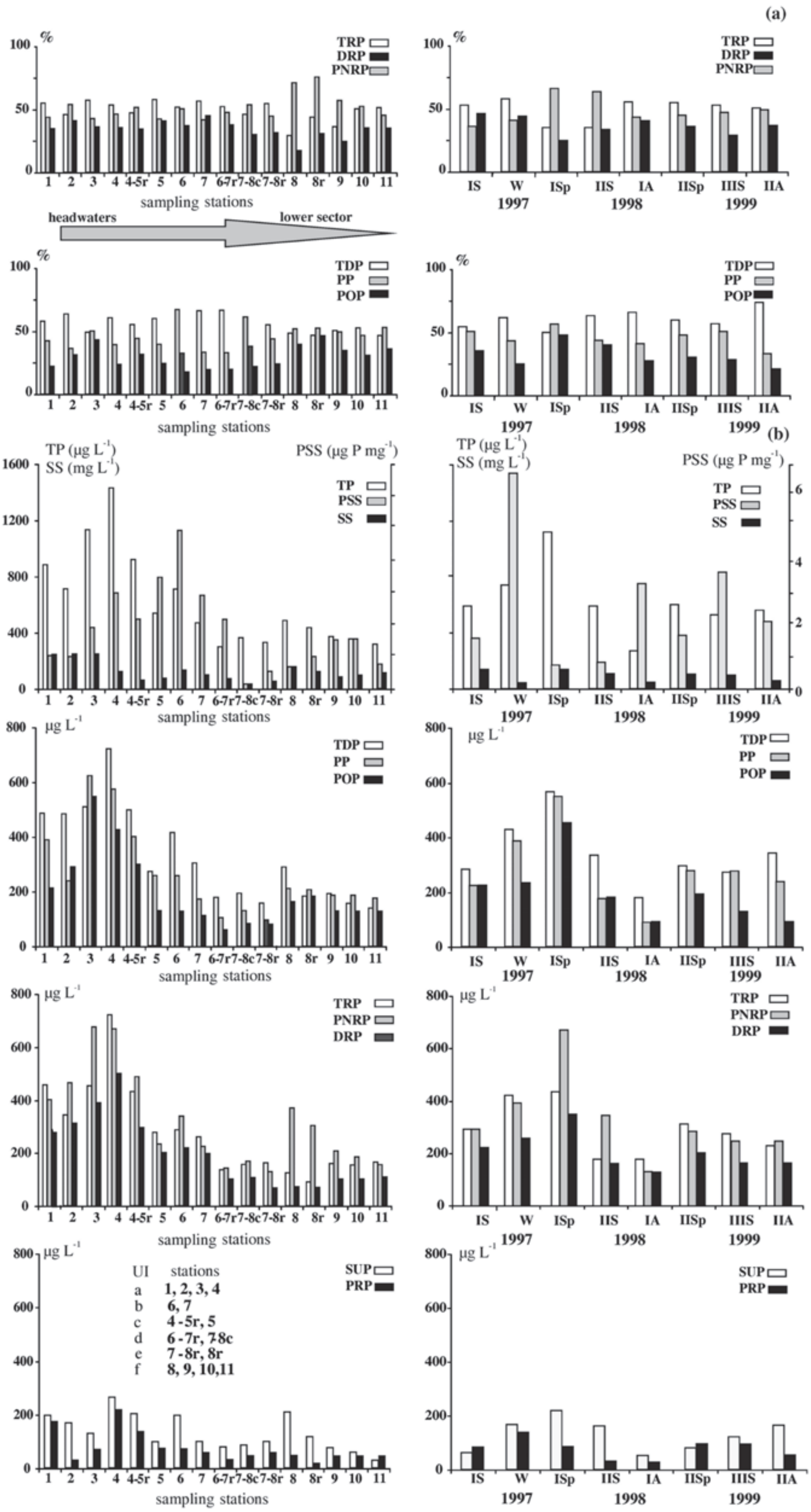

Fig. 3. (**a**) Mean percent contribution of the main fractions of phosphorus (P) to the total P concentration for each sampling station (left panels) and at each sampling occasion (right panels). (**b**) Mean concentrations of the main fractions of P and suspended solids (SS) for each sampling station (left panels) and at each sampling occasions (right panels) (IS: first summer, March/1997; W: winter, July 1997; ISp: first spring, October 1997; IIS: second summer, January 1998, IA: first autumn, May 1998; IISp: second spring, October 1998; IIIS: third summer, February 1999; IIA: second autumn, May 1999).

Table 3. Mean and range (standard deviation in parentheses) of the concentrations of different phosphorus fractions (μg L^{-1} P), PSS (μg mg^{-1}), SS (mg L^{-1}) and OM (mg L^{-1}) in all sampling stations of Salado River basin during 1997–1999 (number of samplings = 8). See Table 2 for the abbreviation of the P fractions. The tributaries are named with the numbers of the stations of the main course located upstream and downstream of each affluent. The letter "r" in the tributaries indicates that the stream is an affluent of the right bank of the river. The letter "c" refers to the artificial condition of the tributary (canal).

Stations	TP	TDP	DRP	TRP	PP	PRP	SUP	POP	PNRP	PSS	SS	OM
1	885 (395)	494 (169)	284 (177)	461 (192)	392 (255)	160 (147)	210 (120)	215 (211)	409 (293)	1.3 (1)	246 (196)	53 (52)
	330–1454	203–713	58–472	87–685	122–800	28–431	51–368	0–521	49–770	0–3	54–656	1–148
2	726 (449)	482 (220)	316 (188)	344 (184)	243 (271)	28 (35)	172 (202)	295 (293)	466 (266)	1.2 (1)	78 (100)	22 (32)
	289–1450	205–724	187–638	187–651	84–726	0–89	0–432	70–713	65–798	0–3	14–256	2–78
3	1137 (866)	514 (314)	381 (274)	455 (331)	623 (621)	75 (71)	133 (138)	549 (564)	682 (593)	2.6 (3)	71 (24)	27 (15)
	302–2708	155–1029	117–898	117–1015	129–1959	0–195	36–459	80–1764	131–1878	0–8	40–103	6–50
4	1421 (953)	771 (463)	508 (384)	728 (546)	654 (537)	284 (365)	166 (234)	425 (478)	681 (585)	3.5 (3)	124 (69)	38 (34)
	488–3150	286–1540	167–1292	246–1589	127–1610	0–908	0–704	0–1362	191–1610	0–9	49–264	2–92
4-5r	937 (1141)	500 (585)	295 (328)	435 (536)	403 (583)	140 (212)	205 (269)	300 (363)	502 (615)	2.7 (3)	71 (35)	20 (19)
	309–3500	186–1799	117–1036	117–1637	71–1701	0–601	14–763	0–1100	48–1863	0–8	38–137	1–55
5	541 (191)	274 (122)	208 (91)	284 (84)	259 (167)	76 (68)	103 (117)	134 (124)	237 (135)	4.1 (4)	78 (75)	26.1 (26)
	246–817	118–423	96–375	175–396	16–505	0–165	6–307	35–392	57–433	0–11	17–240	2–75
6	684 (375)	420 (237)	222 (155)	296 (172)	264 (156)	74 (43)	198 (156)	143 (142)	339 (213)	5.1 (11)	144 (216)	49 (68)
	202–1255	167–802	83–460	120–558	35–453	16–141	55–504	0–342	63–697	0–32	1–656	0–206
7	481 (191)	306 (81)	203 (67)	261 (90)	174 (127)	58 (49)	103 (62)	116 (140)	220 (182)	3.4 (3)	101 (111)	38 (42)
	291–835	230–456	135–333	151–423	43–451	0–131	4–193	26–451	56–644	0–9	9–341	2–117
6-7r	296 (102)	183 (77)	102 (72)	136 (56)	107 (80)	34 (40)	81 (66)	59 (71)	140 (86)	2.7 (4)	70 (57)	23 (18)
	146–431	89–287	36–230	66–230	0.3–209	0–100	25–229	0.4–181	39–264	0–12	4–135	1–48
7-8c	361 (197)	197 (130)	107 (77)	158 (82)	131 (111)	51 (51)	90 (56)	81 (57)	171 (84)	1.8 (2)	39 (30)	20 (7)
	97–566	74–358	21–198	36–214	23–238	15–124	38–160	8–137	61–248	0–4	3 64	12–27
7-8r	332 (199)	148 (82)	64 (49)	162 (108)	92 (97)	98 (95)	60 (37)	80 (141)	129 (150)	1.3 (1)	53 (45)	23 (5)
	58–488	36–230	18–120	21–246	23–240	3–218	18–202	0–291	37–353	1–3	9–100	18–30
8	487 (348)	287 (316)	77 (65)	125 (88)	212 (56)	49 (24)	210 (266)	163 (63)	373 (286)	1.4 (1)	165 (153)	40 (30)
	257–1100	83–846	14–166	36–249	134–259	22–83	44–680	71–220	115–851	1–3	25–406	7–71
8r	421(52)	195 (113)	76 (43)	97 (35)	206 (94)	21 (8)	120 (87)	184 (91)	304 (3)	0.9 (1)	131 (117)	33 (24)
	366–468	123–326	29–111	59–128	103–286	15–30	38–215	87–267	301–307	0–1	0–225	10–58
9	369 (120)	187 (75)	109 (74)	159 (92)	191 (97)	54 (34)	78 (42)	125 (89)	204 (96)	2.2 (2.4)	90 (48)	28 (22)
	182–603	74–270	36–243	45–334	101–405	0–110	27–141	10–295	37–373	0–7	15–174	9–78
10	340 (147)	164 (76)	105 (45)	149 (52)	181 (120)	45 (52)	56 (67)	125 (121)	186 (112)	2.2 (2)	90 (57)	28 (16)
	130–546	89–310	63–198	87–206	23–348	28–81	0–185	5–340	41–340	0–7	11–179	5–56
11	318 (110)	142 (53)	111 (53)	162 (69)	176 (89)	50 (52)	29 (29)	124 (77)	155 (91)	1.5 (1)	120 (78)	33 (31)
	120–431	78–213	59–214	71–262	37–287	0–158	0–63	12–258	32–320	0–5	27–206	1–84

concentrations were detected at stations near the mouth and at St. 2 (IS, IIS, IIIS). The maximum values of particulate unreactive P (PNRP) (> 1600 µg L^{-1}) were determined at Sts 4 and 4-5r, while the minimum was recorded at the river mouth and in the tributaries (LU d and e) (IA). POP concentrations exceeded 1,000 µg L^{-1} at the stations located at the headwaters (LU a and c).

Figure 3 a presents variability in relative contributions of different P fractions among different sampling stations (left panels) or different sampling occasions (right panels). The reactive fractions of P (TRP and DRP) were the major constituents of TP at all sampling occasions except during the spring of 1997 (ISp) and the summer of 1998 (IIS) when the PNRP fraction accounted for up to 50 % of the TP concentration at most sampling stations. For the entire basin, the TDP accounted for 50 % of the TP, with 20 to 30 % representing the bioavailable P (DRP). POP accounted for approximately 25 % of the TP, with the maximum contribution occurring in spring (ISp and IISp).

Patterns in changes of mean P concentrations averaged over different sampling occasions (left panel) or sampling stations (right panels) are presented in Fig. 3b. The mean TP concentrations decreased along the main channel with the lowest value (< 400 µg L^{-1}) being detected at the stations that receive water from LU d and e. Similarly, the mean concentrations of other P fractions [TDP, PP, POP, TRP, PNRP, DRP] tended to be high and low in headwater and tributary stations, respectively. Variability in mean concentrations among stations was relatively large for POP, which displayed the highest values in headwater stations including those associated with shallow lakes (Sts 3 and 4). Downstream from these sites, the values decreased abruptly, and the minima (< 10 µg L^{-1}) were recorded in the tributaries of the middle basin (Sts 6-7r, 7-8c). Seasonally, the mean POP concentration (averaged over sampling stations) was high and low in spring (ISp) and autumn (IA and IIA), respectively. During a low-water (low flow discharge) period in winter at the sampling stations located downstream from major basin cities (Sts 4 and 7), the highest levels of DRP were observed (data not shown). The highest mean PSS were recorded at St. 6 and St. 4. The minimum was measured at St. 8 r. The seasonal maximum occurred in winter, while the minima were detected in the spring of 1997 (ISp) and the summer of 1998 (IIS) with values lower than 1 µg P mg^{-1}. In the middle- and lower-basin sectors (zones d, e, and f), the SUP concentrations were close to those of DRP. The particulate reactive P (PRP) tended to be lower than PNRP at headwater stations.

A comparison between sampling occasions at the time of the two anomalies – the El Niño in the spring of 1997 (ISp) and the La Niña in the spring of 1998 (IISp) – revealed that the concentrations of both the soluble and particulate P fractions were consistently higher during the El Niño than the La Niña period. The mean TP concentration recorded during the former period was about two-fold higher than that recorded during the latter period.

The results of the multiple-regression analysis for all sampling occasions indicated a significant relationship between the P fractions and the LUI. As to the seasons, the summer was a period with no relationship

Table 4. Results of the multiple regression analysis to examine the relationship between concentrations of the major P fractions and LUI, OM, and SS. The analysis was conducted for all the samples as well as for each season (Coefficient not significant: ns). ** $p < 0.01$, * $p < 0.05$. See Table 2 for the abbreviation of the P fractions.

	LUI	OM	SS	Adjusted r^2	Probability
All sampling					
n = 24					
TP	**			0.93	0.001
TDP	**			0.84	0.006
DRP	*	ns	ns	0.82	0.100
PP	**			0.90	0.002
Winter					
n = 6					
TP	*			0.73	0.018
TDP	ns			0.47	0.080
DRP	*		ns	0.88	0.018
PP	*			0.77	0.010
Spring					
n = 6					
TP	**			0.82	0.008
TDP	*			0.70	0.020
DRP	*			0.77	0.013
PP	*			0.74	0.017
Summer					
n = 6					
TP					
TDP	ns	ns	ns	0.67	0.190
DRP			ns	0.09	0.290
PP					
Autumn					
n = 6					
TP	*			0.72	0.020
TDP	ns			0.25	0.180
DRP	*			0.57	0.049
PP		ns		0.25	0.170

Table 5. Results of correlation analysis to examine how the distance from the river mouth is correlated with the concentrations of different phosphorus fractions, SS, and OM. ** $p < 0.01$, * $p < 0.05$. See Table 2 for the abbreviation of the P fractions.

N = 16	Distance	r	Probability
TP	**	0.79	0.000
TDP	**	0.84	0.000
TRP	**	0.79	0.000
DRP	**	0.82	0.000
PP	**	0.67	0.004
POP	**	0.68	0.004
PNRP	**	0.76	0.001
SUP	**	0.70	0.002
PRP	**	0.55	0.026
SS	ns	0.23	0.385
OM	ns	0.18	0.500

between the P fractions and the LUI. Throughout the remaining three seasons the relationships were significant except for the values for TDP during the winters and autumns (Table 4). In this analysis, the concentrations of OM and SS were not significantly correlated with any P fraction.

The distance to the mouth was not significantly related to PM and OM concentrations, whereas it was significantly related to the concentrations of all the P fractions (Table 5).

In the cluster analysis including all the P fractions, the SS and OM concentrations, and the distance to the mouth, two clusters were distinguished. Group I included all sampling stations located at headwaters, whereas Group II was separated in two subgroups: IIa, formed by sampling stations located in the middle basin, and IIb, comprising those situated in the tributaries from the mountains and the lower sector of the basin (Fig. 5).

The results of K-means analysis for Group I consisted in the headwater sampling stations (St. 3 and St. 4), which were linked (lower Euclidean distance) with Group II (Sts 1, 2, and 4-5r). Group II was related to Group IV through sampling stations located in the middle basin (Sts 5, 6, and 7). Group III was composed of| the stations 8 and 8 r and was related to groups IV and V. The last group comprised the sampling stations of the lower basin (Sts 9, 10, and 11) and the tributaries of the sierra sectors (Sts 6-7r, 7-8c, and 7-8r).

Flooding condition in the lower-basin sector

During this investigation, the quarterly records of accumulated rain for 2002 were higher than the historical mean record (1911–2002). The first quarterly record of 2002 duplicated the historical mean in all locations analyzed – those located the nearest to the sampling stations (Dolores and General Belgrano towns) as well as those representing of the headwaters (Junín town) and the sierra subbasin (Azul town). The greatest difference was observed in the record of Belgrano (207 % of the historical mean). The records of the second and third quarters were similar to or lower than the his-

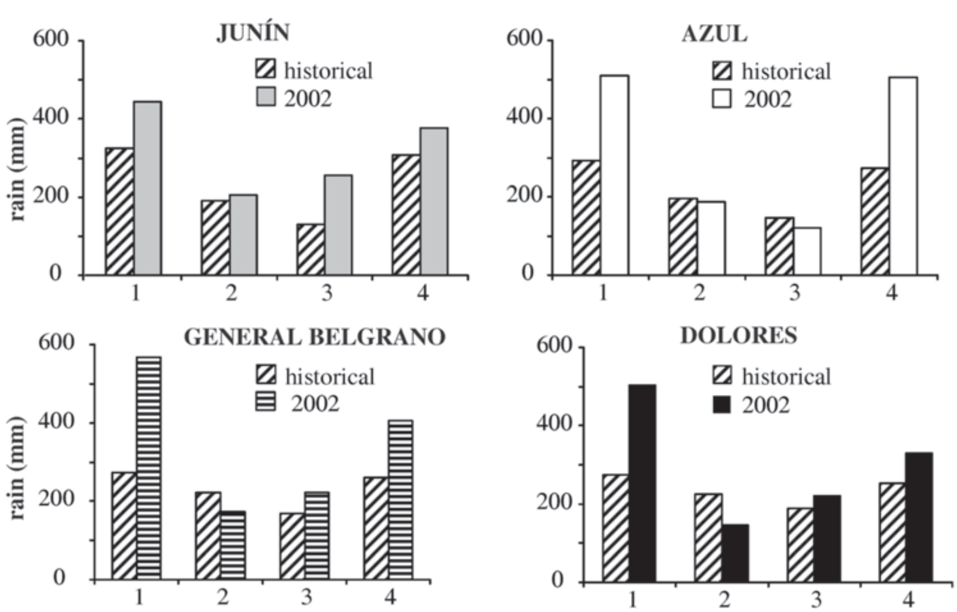

Fig. 4. Comparison between the 2002 mean quarterly rainfall data of localities representative of the subbasins (Junín, Azul, Gral. Belgrano, and Dolores) with historical mean daily rainfall data (1911–2002).

torical mean except for the value for Junín in the third quarter (with a record at 195 % of the historical mean). In the last quarter of 2002, all the records were higher than the historical mean (Fig. 4).

The maximum TP was measured at the stations located at greater distances from the mouth (St. 10), with a clear decrease in values approaching the mouth (St. 12). The range of values was the highest at the stations most distant from the mouth. This trend was similar for the PP concentrations, with the maximum likewise being located at St. 10 and the minimum at St. 12. Nevertheless, the dissolved fraction TDP did not show the same pattern with respect to the distance from the mouth, having a maximum at St. 9 and a minimum at St. 10. The reactive P was higher in the dissolved fraction, with similar values among all the sampling stations. A maximum DRP value, however, was recorded at St. 10 and the minimum at St. 11. The TRP showed the same pattern, with the highest values at Sts 9 and 10 and the lowest at Sts 10, 11, and 12 (Table 6).

The highest mean TP with respect to season was recorded in the spring, mainly corresponding to the PNRP fraction. The TDP and TRP fractions remained unmodified during the sampling period. The maxima for the TRP fraction were recorded during the autumns and the summers. The SUP showed an inverse pattern to that of the TRP, with a maximum during the springs (Fig. 6a).

With respect to the proportions of the different P fractions within the TP, the TDP made the greatest contribution during the winter at 66 %. Within this fraction, the SUP always was present at a higher percentage than the DRP except during the autumn. During the spring, the maximum TP values reflected increases in PP (at 72 % of the TP), mainly in the nonreactive form (PNRP at 85 % of the TP; Fig. 6a, 6b). The DRP was the main form within the PRP (Fig. 6b).

The maximum loads of TP were recorded in Sts 9 and 10 (260 and 194×10^3 kg d^{-1}, respectively) in spring. These coincided with the heavy rains throughout the entire basin that had occurred during the previous week (more than 100 mm). At St. 11, the trend was similar, but with values at less than half of those of the aforementioned. Station St. 12 showed similar TP loads to those observed at St. 11 (maximum at 50×10^3 kg d^{-1}; Fig. 7).

Table 6. Mean and range (standard deviation in parentheses) of the concentrations of different phosphorus fractions in lower sector sampling stations of the Salado River during 2002–2003. Unit is μg L^{-1} P, except for PSS (μg mg^{-1}). See Table 2 for the abbreviation of the P fractions.

	St. 9	St.10	St. 11	St.12
TP	1198 (805) 618–2630	1227 (927) 404–2782	881 (246) 526–1168	850 (257) 434–1321
DRP	200 (33) 174–255	200 (36) 168–275	193 (40) 128–240	185 (20) 159–225
PP	691 (650) 153–1841	817 (920) 123–2440	479 (217) 184–759	398 (220) 92–795
TRP	276 (52) 205–373	272 (65) 174–373	255 (65) 174–348	249 (68) 174–342
TDP	507 (164) 373–789	414 (86) 281–526	398 (56) 342–495	453 (115) 327–618
PRP	76 (55) 0–184	75 (66) 0–184	62 (53) 0–159	64 (61) 0–153
SUP	306 (157) 161–600	217 (80) 113–337	205 (45) 153–290	268 (119) 138–459
POP	616 (641) 37–1657	738(897) 80–2256	421 (219) 139–700	333 (192) 86–657
PNRP	922 (783) 328–2257	955 (897) 193–2409	626 (225) 337–948	601 (223) 260–994
PSS	12 (11) 3–32	13 (13) 3–35	8 (4) 4–14	6 (2) 2–9

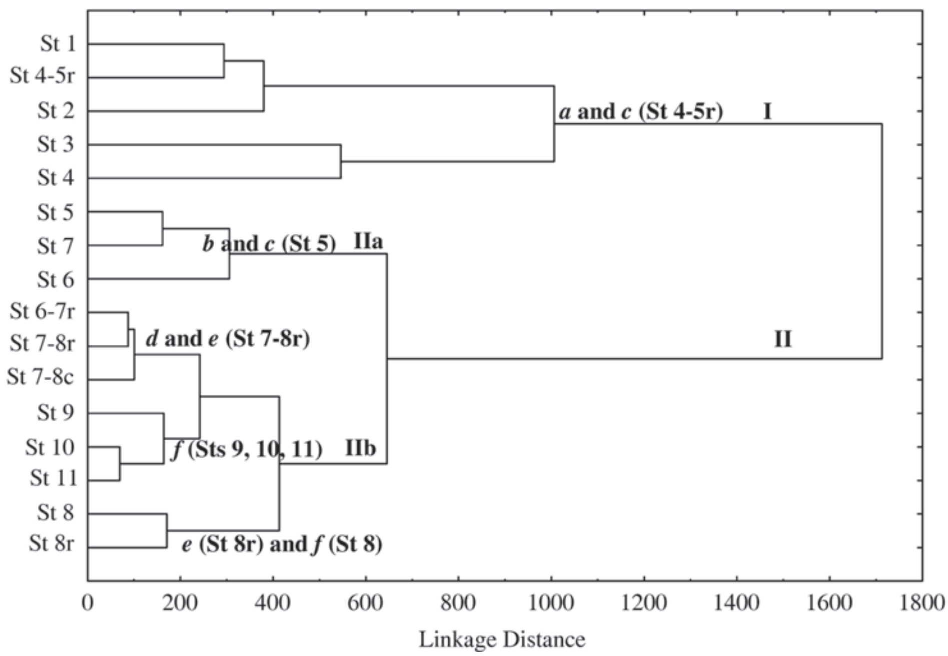

Fig. 5. Grouping of samples by cluster analysis according to the concentrations of different phosphorus fractions, organic matter, suspended solids, and the distance from the river mouth. Letters in italics indicate the land use units. Roman numerals with lowercase letters show the main groups identified by the cluster analysis.

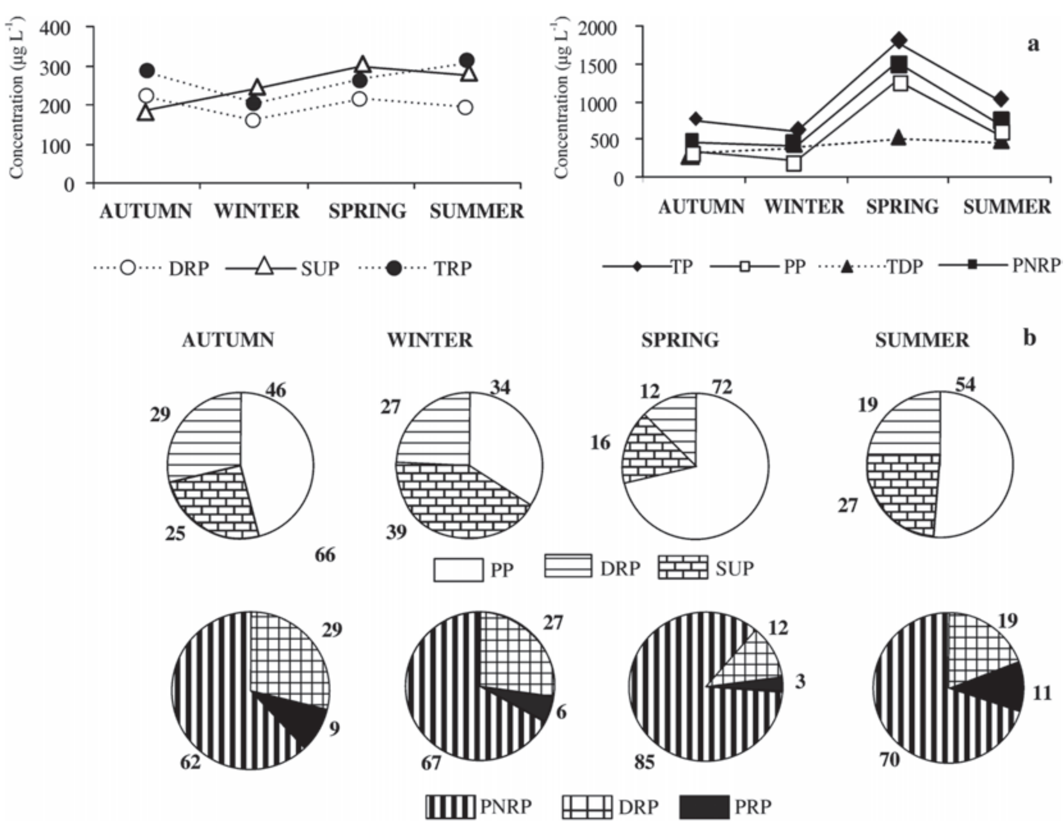

Fig. 6. (a) Seasonal mean-concentration values for different phosphorus fractions for all sampling stations during the flooding period (2002–2003). **(b)** Contributions of the different phosphorus fractions to total phosphorus during the same sampling period.

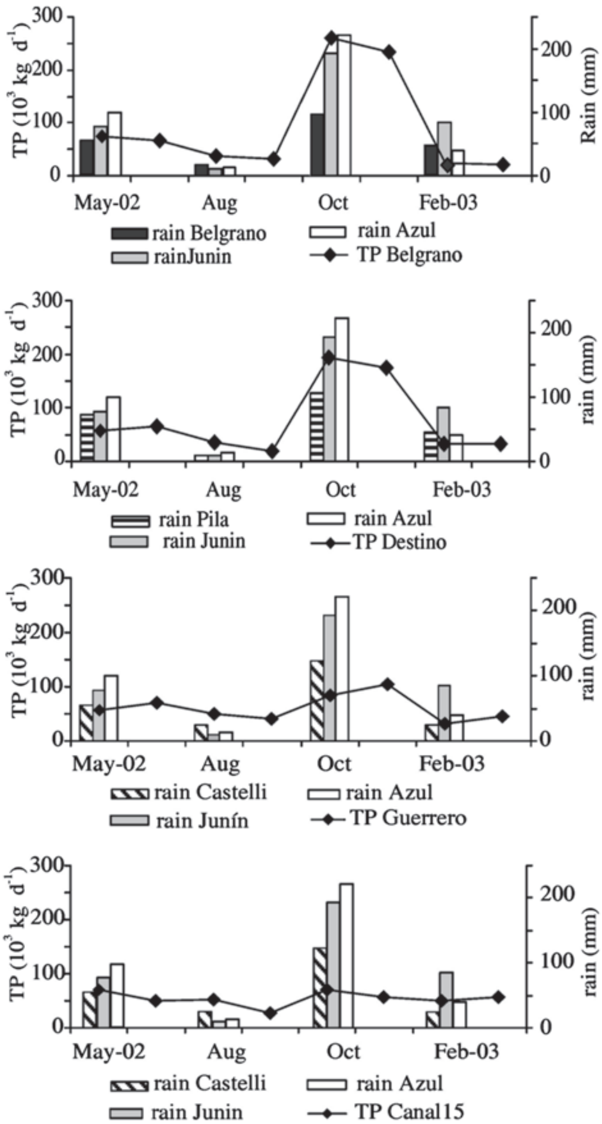

Fig. 7. Comparison between local and subbasin rainfalls (for Junín and Azul cities) along with seasonal total-phosphorus load for each sampling station during a flooding period (2002–2003)

The results of ANOVA indicated that the differences in the P loads between first and second sampling days at the four sites of the lower river sector during the flooding condition (May 2002, August 2002, October 2002, February 2003) were not significant ($n = 32$ $p = 0.82$).

Discussion

Regional sampling

The sampling period began during a major El-Niño period (1997–1998) and ended in a La Niña event (1998–1999; Nieto Ferreira et al. 2003). These meteorological changes could influence the crop yield on a local scale (Podestá et al. 1999, Bettolli et al. 2009). The global analysis of soybean production related to the ENSO multivariate index (ENSO MEI) and related rains, however, showed a continuous growth from the beginning of the 1990s (Fig. 2). The maxima of the P fractions were registered during the spring of 1997, whereas in the spring of 1998 the hydroclimatic conditions – the rainfall in sectors LU a and LU c being less than half that in 1997 – could explain the decrease in TP concentration (Gabellone et al. 2005). Nevertheless, the levels of TP recorded in the spring 1998 were also the highest for the period 1998–1999. The increased production of soybean implied an enhanced application of fertilizer that, along with the increase of nonpoint sources of P, would ultimately enter the aquatic environments (Carpenter et al. 1998). Since the Salado-River basin is a major watershed with extensive agricultural use, this situation would imply an increase in P levels in the water. On the basis of the mean values of TP and TDP estimated for the entire basin of the Salado River, this body of water must be considered eutrophic (a heavily enriched river; Dodds et al.1998, Environment Agency UK 1998, Alexander & Smith 2006). The highest values of PSS also indicated the presence of point and nonpoint sources of P (Edwards & Withers 2007). The distribution of the different forms of P in the basin is not, however, uniform: rather, a clear gradient is present involving a decrease in P concentrations toward the mouth (Fig. 4). These spatial changes were related to the diverse LU as well as to the agricultural calendar, a finding consistent with the results of Edwards & Withers (2007) and Jarvie et al. (2010) in European basins.

The diversion of the water from an endorheic catchment through artificial canals into the Salado River during the last decade is another condition favoring the increase in the nutrient loads into the Salado basin. This form of connectivity has been emphasized by Edwards & Withers (2007) as a cause of nutrient enrichment in rivers. The P input from the Saladillo stream (St. 4-5r) comes mainly from a sand-dune–containing sector (LU c). The values of TP recorded at this station were the highest in the basin (3,500 µg L^{-1}). The other locations with high levels of TP were situated at the headwaters (LU a), there characterized by intensive agricultural activity.

PP was the main fraction in the headwaters (LU a, with intensive agriculture) in agreement with the results of Hodgkinson & Withers (2007) for UK rivers.

The input from the tributaries of the middle sector of the river (LU d and e) with low P loads – coupled to an increased discharge and a change in LU, where agriculture was replaced by extensive cattle-raising – all effected a decrease in P in the main canal. The abundance of phytoplankton (Neschuk 2001) and the presence of lentic environments associated with the river (e.g., with backwater ponds and flushing lakes) could have played a consequential role in producing the diminution in P (mainly in the soluble fraction) in the middle sector of the river. These environments can be considered as a service ecosystem, whose conservation is critical in avoiding the eutrophication of the rivers (Carpenter et al.1998).

The DRP constituted the main fraction of the TDP and was furthermore found to be the predominant P form in the headwaters in accordance with the bioavailable reactive fractions, there of great ecological relevance (Edwards & Withers 2007). During the low-water period (winter) the highest values of DRP were recorded at the sampling stations located downstream from the major towns. This condition could be related to the inputs from point P sources because during this season the agricultural activities are only minor (Jarvie et al. 2008, Oberholster et al. 2013). In the middle and lower sectors of the basin (LU d, e, and f), the nonbioavailable TDP fraction (SUP) exhibited similar proportions of DRP. The bioavailable fraction of the PP, the PRP, was lower than the nonreactive fraction (PNRP), mainly at the headwaters. Consistent with the results of Edwards & Withers (2007) for rural rivers, the TDP of the Salado always constituted 50 % of the TP, whereas the DRP, in turn, represented some 20–30 % of the TDP.

Flooding condition in the lower-basin sector

During an extended flooding, the peak of TP was observed during the spring, coinciding with the maximum rainfall recorded in the basin and related to periods with the highest agricultural activities (Neal et al. 2005, Jarvie et al. 2010). The maximum P levels recorded in October 2003 were related to an increase in PP (Obermann et al. 2007), mainly in the form of PNRP. The other P fractions underwent small modifications. Nevertheless, the maximum for all the P fractions during this recurring flooding period (about 10 years after the previous one) were higher than those recorded, at the same sampling stations, in the earlier interval (1997–1999), when at that time the basin had exhibited medium and low discharges.

In October, a peak of P loading was detected along with a clear decrease in the P load at the lower-basin stations to the mouth. This condition could be related to the presence of influential flushing lakes between El Destino (St. 10) and Guerrero (St. 11) and also the existence of wetlands located between Guerrero and Canal 15. On the other sampling occasions, such decreases were not detected since the entire lower-basin sector exhibited similar values of P, especially with respect to the dissolved fractions. The increase in wetland areas might have produced a significant impact on the decrease in the P load transported by the river because the particulate fractions of P become progressively deposited in the wetlands after the spring rains (i.e., the cause of the October P loading) as function of the distance from the headwaters.

Conclusions

In this investigation, we identified the main sources and delivery of P on a large scale, and the relative proportions of the various P fractions under different hydrologic conditions.

The spring peaks of TP corresponded to periods of highest agricultural activities. After the extended flooding of 2002, the spring TP peak coincided with the maximum rainfall recorded in the basin. The maxima for all phosphorus fractions during the second period (2002–2003, a recurrence of 10 years) were higher than those recorded in 1997–1999 at the same sampling stations, when the basin had exhibited medium and low discharges. Nevertheless, in both periods, the considerable decrease in the P load observed near the mouth could be related to the sizeable wetlands present.

In spite of the growth in agricultural production within the Salado-River basin, the mechanism and extent of P transport from agricultural lands to the moving water of the Salado River are still unknown. A worse trophic state of the bodies of water is still possible, which circumstance would exert a clear impact on the so-called RAMSAR site in Samborombón Bay (Ramsar Convention Bureau 2004), since the shallow Samborombón estuary would be as highly sensitive to eutrophication as a typical lake system. A public environmental policy for controlling the agricultural P sources and a coalition of different sciences such as agronomy, hydrology, and limnology are all necessary in order to produce a truly integrated approach to the problem of the eutrophication of the Salado River basin since that process has been shown to have significant impact on both economy and public health.

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