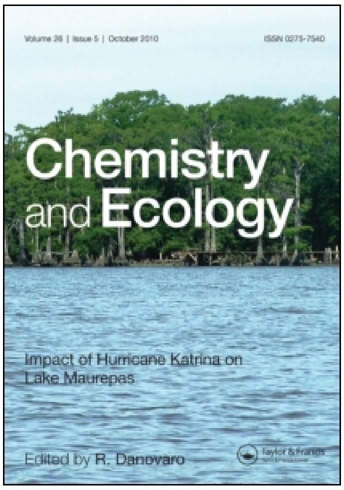

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THE CHEMICAL COMPOSITION AND FLOW OF THE RIVER LEA, SPAIN

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Weekly measurements of chemical composition were made at four sampling sites on the Lea River (Spain) between July 1985 and August 1986. At two stations there were gauging-weirs for stream discharge monitoring. Conductivity, pH, alkalinity, silicon, calcium, magnesium, sodium, potassium, ammonia, nitrate, sulphate, chloride, reactive phosphate, and total phosphate, were recorded in each site.

Results were analysed using multiple regression techniques to study the relationship between flow and chemical components.

Conductivity, alkalinity, calcium, magnesium and sulphate were strongly related to flow, decreasing in concentration with increased flow. Ammonia, potassium, reactive phosphate and total phosphate varied most widely, with peaks at times of heavy rainfall. Nitrate increased at times of high discharge, with little variation between sampling points. Silicon decreased in summer and autumn in the lower catchment area whilst chloride and sodium were higher in the lower catchment of the River Lea.

KEY WORDS: Chemical composition, flow, spatial variation, seasonal variation, Spain (River Lea).

INTRODUCTION

The high degree of environmental damage in the Basque Country, specially of the rivers, has prompted many hydrological studies (Gobierno Vasco, 1986; Arluciaga and Alzate, 1984; Azcona *et al.*, 1984; Bikuña *et al.*, 1987; Gómez *et al.*, 1987), with the aim of establishing a water quality classification for administrative action.

This paper presents results which are part of a comprehensive environmental research programme for Atlantic seaboard rivers of the Basque Country. This study will provide basic information required for the development of recreational fisheries and the monitoring of the biological recovery processes during the restoration of polluted rivers.

The results of the chemistry study are based on an intensive one-year sampling of the River Lea to determine seasonal changes and the effect of floods in chemical transport.

STUDY AREA

The River Lea (see Fig. 1) is a short stream, 22 km long, in the Bizkaia Province (Basque Country, Spain). Its source is on the north slope of Mt. Oiz (1026 m), and it

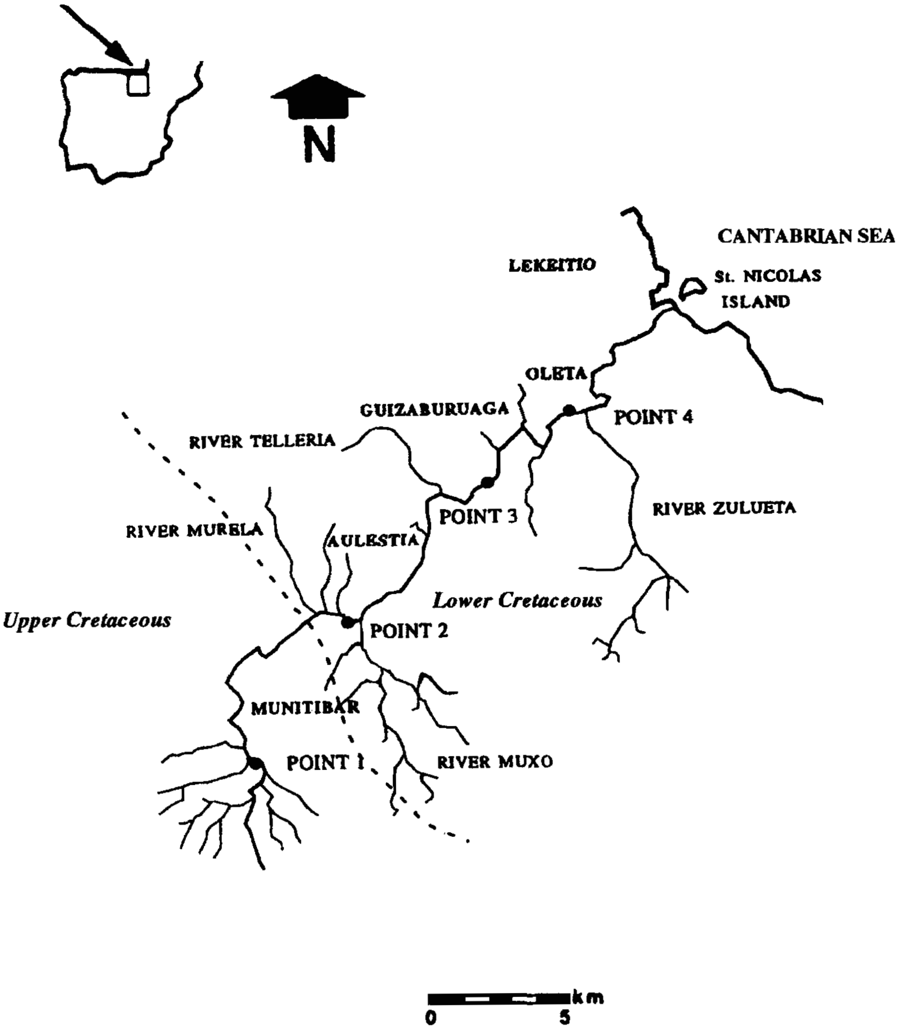

Figure 1 River Lea map, showing the tributaries and sampling points.

flows into the Cantabrian Sea at Lekeitio. Its catchment has an area of 80 km^2, rising in the hills to a height of 300 m with a slope of 13.6 m km^{-1}. The Lea has many small tributaries, such as the Rivers Zulueta, Telleria, Muxo and Murela. An exhaustive morphometric study is presented in Eraso (1983).

The Lea catchment has a very low population (0.24 people ha^{-1}), whose main activity is farming. There is no major industrial activity in the catchment.

There are two well defined areas of geology, Upper Cretaceous, with limestones, shales and siltstones in the upper catchment above Aulestia; and Lower Cretaceous, with clays, sands and Urgonian limestones in the rest of the catchment to the river mouth.

The River Lea can be defined as a low flow river, with a torrential regime depending upon the rainfall, the discharge showing a very fast response. Monthly average rainfall between 1974 and 1984 was 129.4 mm m^{-2}, with a montly average temperature range of 24.7° C to 2.3° C (Sondica Airport Metereological Station, pers. comm.).

The choice of sampling points was made taking into account the distance to the river sources, altitude and geology (see Tab. I).

MATERIALS AND METHODS

Water samples were taken weekly (on Monday) from the described points (see Fig. 1), through a one-year period beginning in July 1985 and ending in August 1986. One litre acid-cleaned polyethylene bottles were used to take the samples.

Temperature, pH and conductivity were measured in the field. Water samples were taken back to the laboratory and filtered through pre-washed glass fibre filter GF/C Whatman (4.7 cm, 1.2 nm) (Casey and Walker, 1983), and stored in acid-cleaned polyethylene bottles. Laboratory analysis of samples was generally completed within 24 hours. Alkalinity, silicon, ammonia, reactive phosphate, total phosphate, chloride, nitrate sulphate, calcium, magnesium, sodium and potassium were determined using chemical methods described below:

pH: was measured on a Hanna HI-8424 field pH-meter with temperature compensation, standardized with pH 7.00 and pH 10.00 standard buffer solutions.

Conductivity: was measured on a Hanna HI-8333 field conductivity meter, calibrated with 0.01N KCl standard solution. Measurements were made at ambient temperature, transformed later to 25° C (American Public Health Association, APHA, 1980, Standard Method 205).

Alkalinity: Water samples were titrated with 0.01N HCl standard solution, using mixed bromcresol green-methyl red indicator solution pH 5.2–4.6 (APHA, *loc. cit.* Standard Method 403).

Ammonia: Phenol-nitroprusside method after modification of Chaney and Marbach (Mackereth *et al.*, 1978).

Silicon: Soluble reactive silicon, Heteropoly Blue Method (APHA, *loc. cit.* Standard Method 425 D).

Table I Description of sampling points.

Sampling point	*Slope m km^{-1}*	*Distance from source km*	*Altitude m*	*River bed*
1. Guerricaiz	66	4.0	240	Solid rock and boulders
2. Aulestia	10	9.5	100	,,
3. Guizaburuaga	10	16.0	40	Boulders and sand
4. Oleta	10	18.0	20	,,

Reactive Phosphate: Ascorbic acid method on filtered sample. (APHA, *loc. cit.* Standard Method 424 F).

Total Phosphate: Ascorbic acid method with preliminary digestion step for total phosphorus (APHA, *loc. cit.* Standard Methods 424C III).

Calcium, Magnesium, Sodium and Potassium: Measured using a Perkin-Elmer atomic absorption spectrophotometer, model 2280).

Chloride, Nitrate and Sulphate: Ion Chromatography by High Performance Liquid Chromatography (HPLC). Pump: Waters Model 510. Injector: Water Model U6K. Absorbance detector: Water Model 440. Wavelength: 254 nm. Column: Anionic Vidac 302 IC.

Suspended solids were not determined during the present study due to their low levels in normal flow conditions.

Discharge records at site 2, Aulestia, and site 4, Oleta were provided by the Diputación Foral de Bizkaia, and the precipitation data by the Sondica Airport.

Statistical analysis: A general statistical analysis of chemical data was carried out using Principal Component Analysis, without rotation, on untransformed variables (Cuadras, 1981).

The description of physical and chemical variables was by means of a model derived by regression analysis (Snedecor and Cochran, 1967), including point and season information to modify the coefficients of equations (Draper and Smith, 1966). The objective is to obtain the simplest and best fitting models, according to the following scheme:

1. Study of the basic relations between flow and water parameters. Two models were considered, choosing in each case the best fitting model:
   - Linear regression:

$$Y = a + bX$$

   - Logarithmic regression:

$$\log_e Y = a + b \log_e X$$

2. Modification of the coefficient a to take into account both sampling point and/or seasonal influence. The actual values of this coefficient are obtained in each case by summation of data included in Table III.
3. Modification of the coefficient b to take into account the influence of sampling point on flow. Actual values are again obtained in each case by summation of data in Table III.
4. To accept any modification of coefficients, the usual contrast of differences between means based on Student's t distribution was used calculated from standard deviation and correlation coefficient. If acceptable, data are divided into two lots (1 and 2) with different regression coefficients.
5. The significance of equations obtained was calculated by Pearson's correlation coefficient. All equations included in Table III have a confidence level greater than 99 percent, because the critical value of r is 0.354 for 50 degrees of freedom.

RESULTS AND DISCUSSION

Rainfall and Flow

Atmospheric precipitation between June 1985 and August 1986 fell always as rain; information was provided by Sondica Airport Meteorological Station approximately 50 km from the Lea catchment. Comparing the daily rainfall means between the periods 1985–1986 (2.95 mm m^{-2}) and 1974–1984 (4.30 mm m^{-2}), the sampling period corresponded to a very dry year and as shown in Figure 2, the summer season extended to the middle of November.

Flow data for this study came from two gauging-weirs: at point two, Aulestia, and at point four, Oleta (Diputación Foral de Bizkaia, pers. comm.).

An attempt was made to find a relationship between flow and rainfall data. The relationship was not close, showing a rain regime localized to the Lea catchment. This phenomenon has been reported by other authors for different areas (e.g. Lopez Unzu, 1986). Consequently, the rainfall data available may not represent rainfall in the Lea catchment.

The flow data from the two gauging-weirs gave the following relationship:

$$Q_4 = aQ_2^b$$

were Q_4 is the flow at point four, Q_2 the flow at point two and a, b, are constants.

This kind of relationship suggests a contribution from both tributaries and runoff to the flow in the main stream, thus producing a modification of the linearity, specially at high flows.

Since the relationship between flows can be adjusted to a straight line and there are no flow measurements for the other two sampling points, all the relationships will be expressed with regard to the flow at the upper sampling point, Aulestia.

Chemistry

The geological and physical features of the area under study and the limited farming and domestic activities in the catchment causes the water of the River Lea to undergo little chemical change along its course. Principal Components Analysis of the data revealed the features of the sampling, with the first axis showing a seasonal influence and the second being explained as a longitudinal gradient (Tab. II).

The only variations are due to (a) dilution because of rainfall, (b) biological processes such as the uptake of silicon by diatoms, and (c) the input of small amounts of sewage effluent.

Seasonality appears as the factor contributing the largest variability, and is expressed mainly through the flow variable, but in some cases a modification of the independent term in equation was also necessary to explain seasonal changes.

Similar behaviour can be observed in several groups of physical and chemical variables. This is attributed to seasonal and spatial variations. Conductivity, alkalinity, calcium, magnesium, potassium and sulphate (see Figs. 3, 4, 5) all show a reduction in concentration during the winter-spring period and an increase during the summer-autumn period. With the exception of conductivity (see Tab. II), all

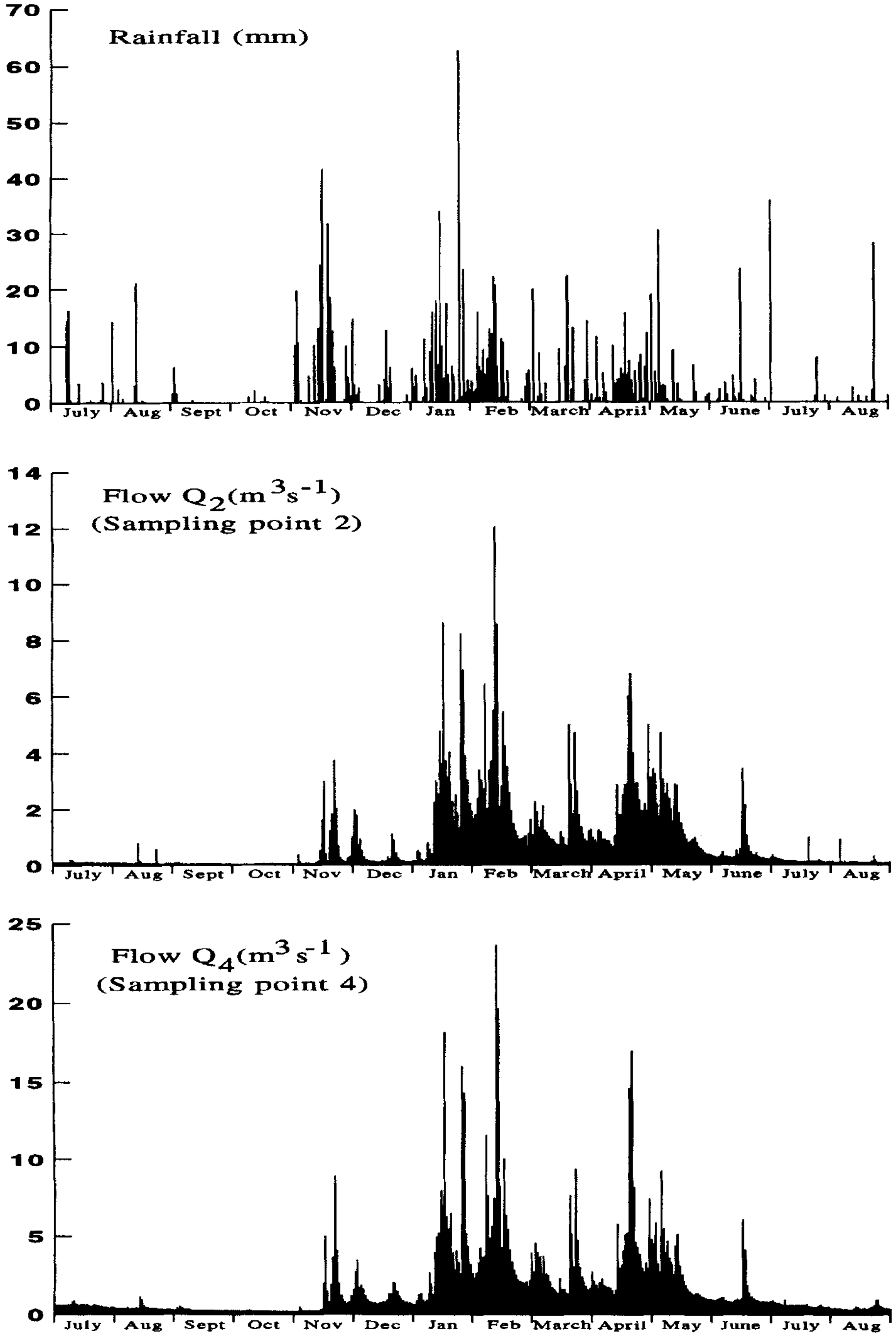

Figure 2 Rainfall (mm) at Sondica Airport; and flow (m^3s^{-1}) from the River Lea at Aulestia (point 2) and Oleta (point 4) gauging weirs.

Table II Summarized chemical and flow data from the four sampling points of the River Lea (max., min. and mean values) over the period July 1985 to August 1986.

		Guerricaiz Point 1	*Aulestia Point 2*	*Guizaburuaga point 3*	*Oleta Point 4*
Flow	max.		12.03		23.62
$m^3 s^{-1}$	min.		0.04		0.19
	mean		0.93		1.87
pH	max.	8.68	8.62	8.29	8.44
	min.	7.83	7.72	7.24	7.68
	mean	8.34	8.18	8.02	8.07
Conductivity	max.	396.00	372.00	398.00	423.00
$\mu S\ cm^{-1} s^{-1}$	min.	179.00	173.00	212.00	245.00
25° C	mean	325.55	303.95	320.18	339.62
Alkalinity	max.	3.86	2.96	2.90	2.97
meq l^{-1}	min.	1.18	1.15	1.54	1.80
HCO_3^-	mean	2.90	2.39	2.43	2.54
Calcium	max	68.67	94.40	58.16	61.16
mg l^{-1}	min.	27.83	21.00	30.00	35.00
	mean	55.28	46.96	47.39	49.81
Silicon	max.	3.27	3.91	2.62	2.40
mg l^{-1} Si	min.	1.33	1.66	1.02	0.95
	mean	2.39	2.60	1.94	1.76
Sulphate	max.	14.75	28.45	23.65	22.01
mg $l^{-1} SO_4^{2-}$	min.	6.82	9.58	10.60	10.43
	mean	9.96	17.29	16.37	16.00
Magnesium	max.	3.36	4.45	4.61	4.93
mg l^{-1}	min.	1.30	1.70	1.60	1.60
	mean	2.34	3.32	3.05	3.07
Chloride	max.	16.39	19.12	28.94	34.14
mg l^{-1}	min.	5.74	7.27	8.03	9.14
	mean	10.15	12.04	15.66	18.60
Sodium	max.	8.67	15.33	15.83	17.33
mg l^{-1}	min.	4.33	5.67	5.16	6.16
	mean	6.33	8.08	9.61	10.90
Nitrate	max.	1.58	1.40	2.00	2.29
mg l^{-1}	min.	0.20	0.12	0.21	0.12
NO_3^-. N	mean	0.76	0.70	0.68	0.76
Total Phosphate	max.	365.60	106.00	107.00	210.00
$\mu g\ l^{-1}$	min.	3.40	5.40	7.20	12.00
PO_4^{3-}. P	mean	23.55	30.93	32.86	40.94
Reactive Phosphate	max.	167.40	34.40	38.00	38.00
$\mu g\ l^{-1}$	min.	0.50	1.20	1.20	1.40
PO_4^{3-}. P	mean	8.89	15.54	14.32	17.39
Ammonia	max.	290.00	152.50	148.50	107.50
$\mu g\ l^{-1}$	min.	3.40	2.20	5.60	2.50
NH_3. N	mean	38.84	29.56	29.12	29.19
Potassium	max.	4.59	4.43	3.04	3.40
mg l^{-1}	min.	0.49	0.56	0.79	0.87
	mean	1.07	2.10	1.84	2.01

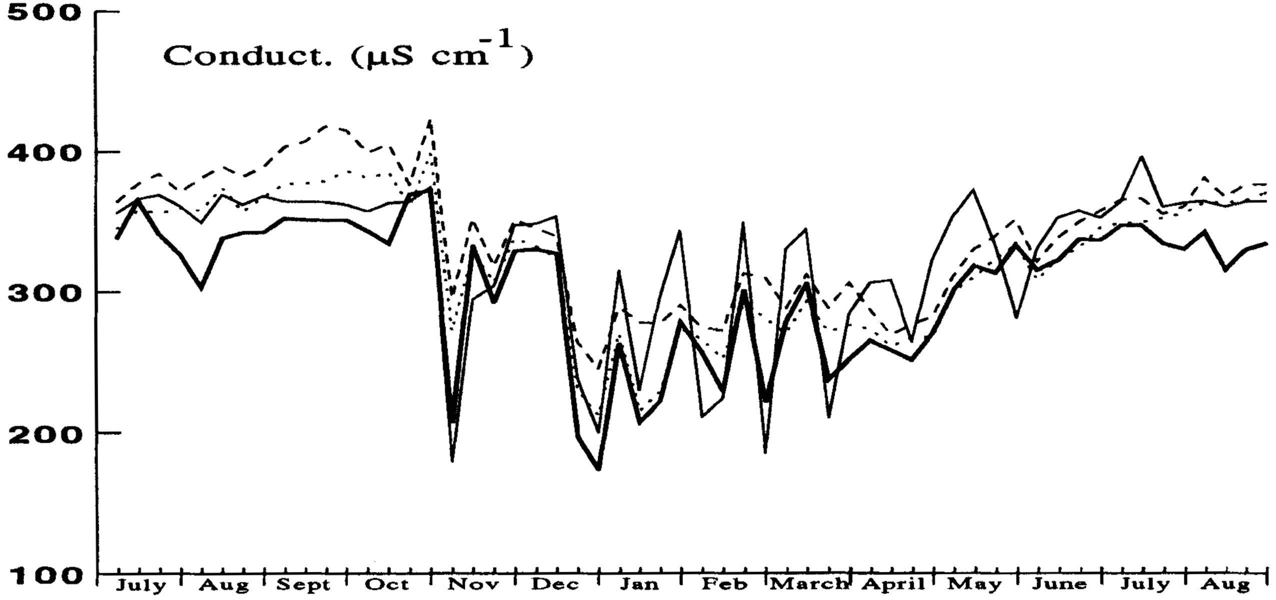

Figure 3 Seasonal variation in conductivity (µS cm^{-1}, 25° C.) for the different sampling points. (__ Guerricaiz; ▬ Aulestia; …… Guizaburuaga; _ _ _ Oleta).

these variables have different values between sampling points 1 and 2. These differences are due to the changing geology of the catchment, the upper basin being more calcareous than the middle and lower basins. Tributaries that flow through terrain overlying Lower Cretaceous rocks and enter the main stream just above the second sampling point (Aulestia) cause a fall in calcium (15%) and alkalinity (17.6%), and an increase in sulphate (73.6%), magnesium (41.9%) and potassium (96.3%).

This observation and the behaviour of each variable with regard to flow are expressed in a different way in each of the mathematical models (see Tab. III). A general model cannot be derived for potassium, magnesium and sulphates unless the first sampling point is considered separately. Sulphate concentrations follow the least general model, showing a complete lack of relation with flow at the first sampling point (Guerricaiz), with very constant values through the sampling period.

Calcium and alkalinity have been adjusted to the general model because they were less variable with regard to flow. These two variables show different relations with flow in the lower stream (points 3 and 4). In all cases, the changes in concentrations along the river are explained by mean variations of the sampling point.

Silicon (see Fig. 6) can also be represented by a general model, without large significant differences between times of the year. However, a dilution effect occurs during the winter ($a_{w_t} = -0.2596$). Along the course of the river the silicon-flow relationship undergoes a gradual change, which can be seen in the interaction term (b_2, b_3, b_4), going from negative coefficients in the upper river to positive ones at the

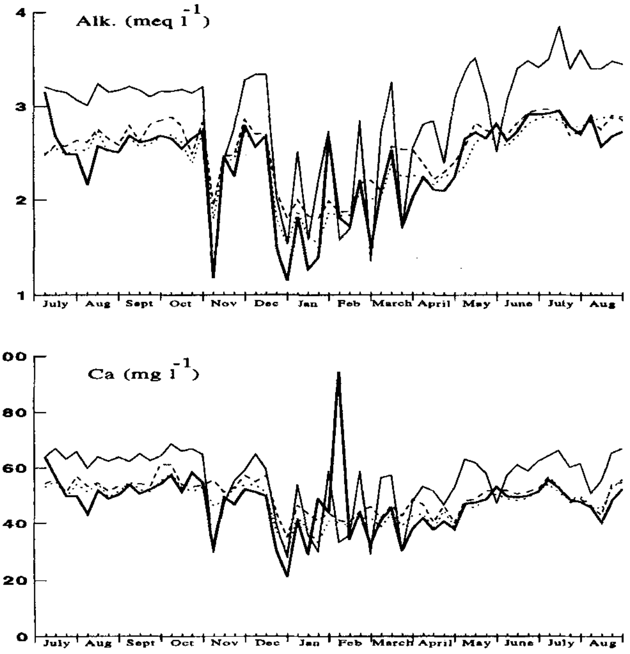

Figure 4 Variation of alkalinity (meq l^{-1}) and calcium (mg l^{-1}) in the River Lea. (—— Guerricaiz; —— Aulestia; ...... Guizaburuaga; — — — Oleta).

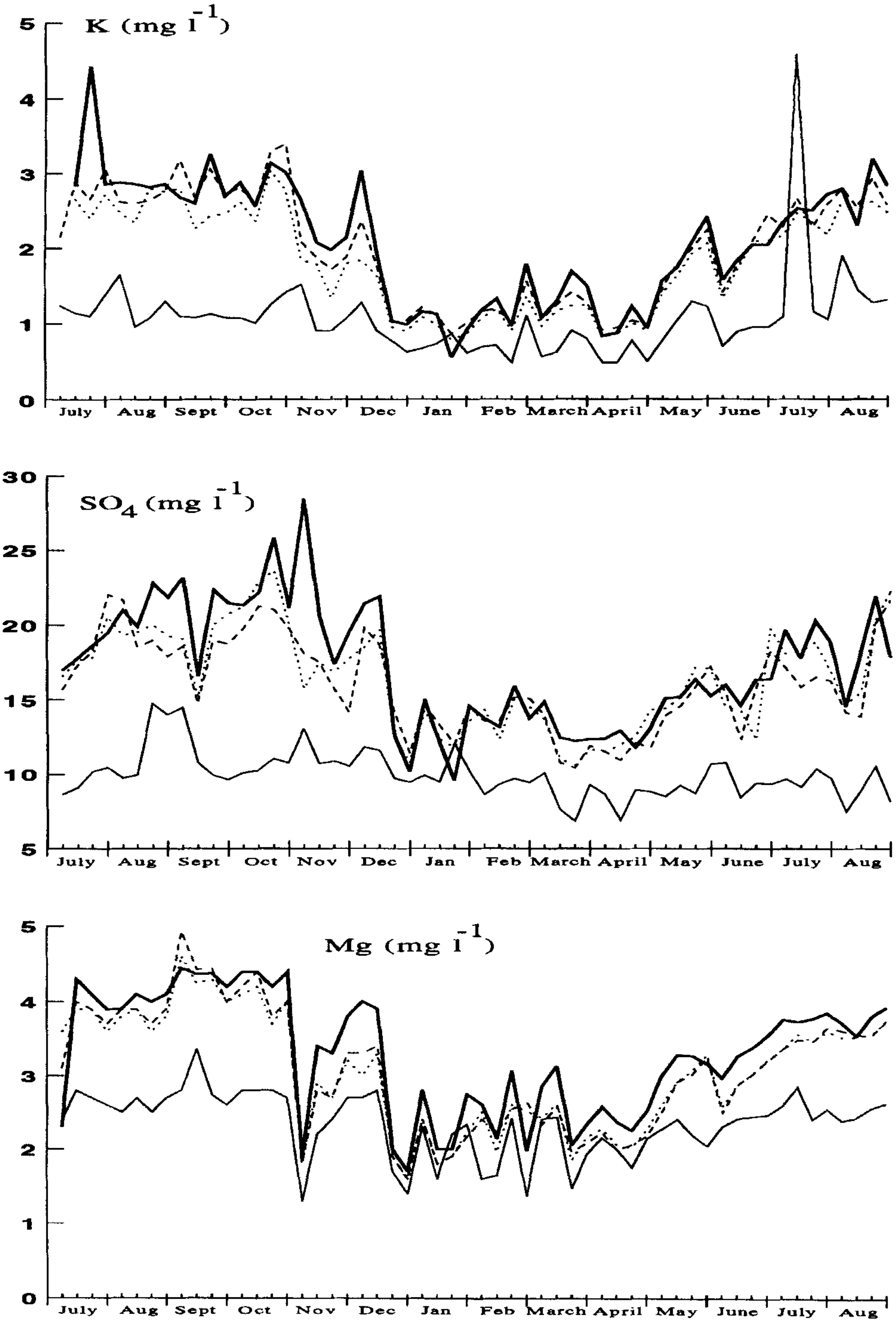

Figure 5 Variation of potassium (mg l^{-1}), sulphate (mg l^{-1}) and magnesium (mg l^{-1}) in the River Lea. (——— Guerricaiz; ━━━ Aulestia; ...... Guizaburuaga; — — — Oleta).

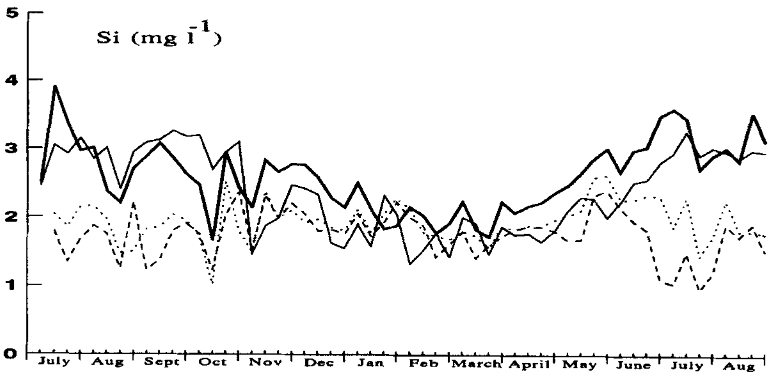

Figure 6 Seasonal variation in silicon (mg l^{-1}) data for the different sampling points. (____ Guerricaiz; ____ Aulestia; ...... Guizaburuaga; _ _ _ Oleta).

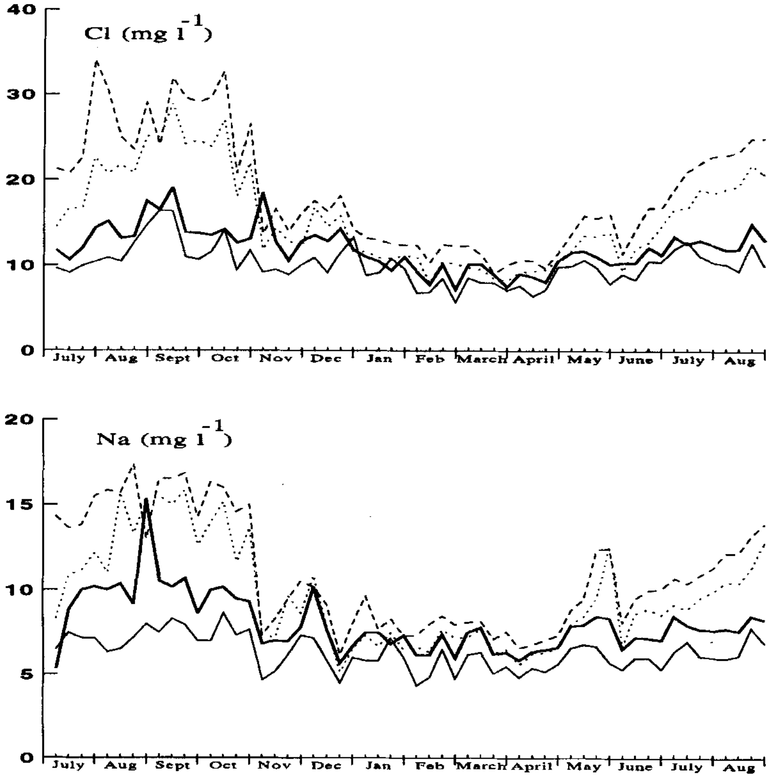

Figure 7 Comparison of chloride (mg l^{-1}) and sodium (mg l^{-1}) data for the different sampling points. (____ Guerricaiz; ____ Aulestia; ...... Guizaburuaga; _ _ _ Oleta).

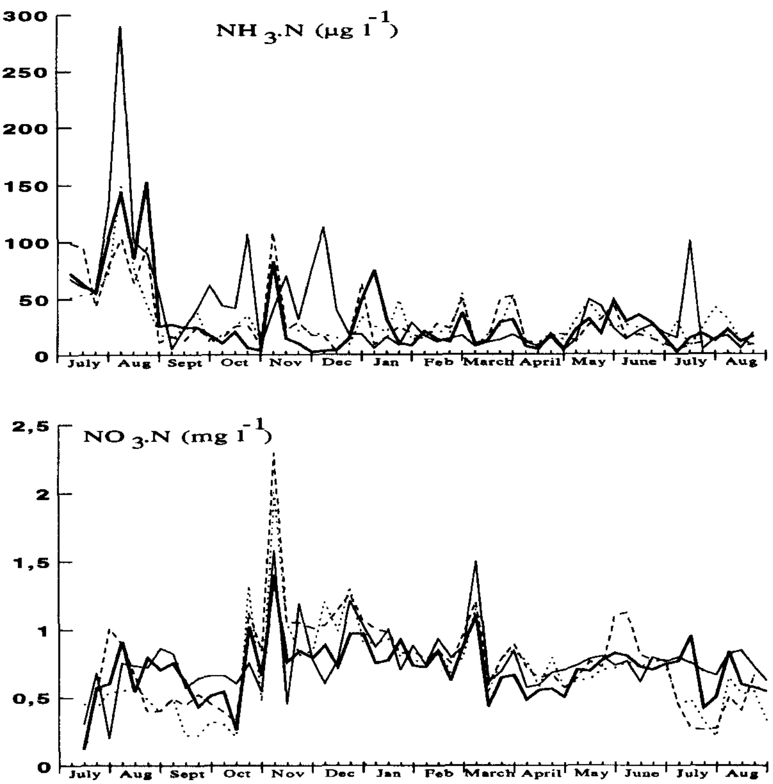

Figure 8 Variation of ammonia (μg l^{-1}) and nitrate (mg l^{-1}) in the River Lea. (____ Guerricaiz; ____ Aulestia; ...... Guizaburuaga; _ _ _ Oleta).

river mouth (see Tab. III). This could be due to diatom growth, with progressive silicon uptake by these microorganisms in the middle and lower sections of the River Lea at times of low flow when the residence time of the water increases (Casey *et al.*, 1981; Saad and Abbas, 1985).

Besides the geological features of the area and biological activity, there are other factors that may influence the quality of the river water, such as inputs of atmospheric origin or sewage effluents. This is the case with chloride and sodium concentrations, whose variations are very similar (see Fig. 7). For both ions, an increase is observed in the concentrations of these elements with proximity to the river mouth (83% Cl, 72% Na). There are differences in the coefficients of the flow in the model for sodium between sampling points 3 and 4 ($b_3 = b_4 = -0.079$) with regard to sampling points 1 and 2 ($n = -0.051$).

For chloride, this effect is so strong that a general model cannot be formulated. This difference between points can be explained by the maritime influence, and also by the presence of small rural populations discharging their sewage to the River Lea. The input of chlorides and sodium, observed mainly in periods of low flow (summer-

Table III Summary of multiple regression analyses of the chemical variables.

VARIABLE	a	a_{p2}	a_{p3}	a_{p4}	a_{S_p}	a_{S_m}	a_{Aut}	a_{W_t}	b	b_2	b_3	b_4	r
Si	2.766		-0.809	-1.058				-0.260	$-2.481 \cdot 10^{-4}$	$1.188 \cdot 10^{-4}$	$2.568 \cdot 10^{-4}$	$3.088 \cdot 10^{-4}$	0.759
LCond.	6.298	-0.059		0.055	0.033				-0.092				0.869
NO_3.N	2.267				0.627		1.438	0.627	$2.403 \cdot 10^{-4}$				0.558
AlK	3.136	-0.530	-0.530	-0.530	0.227	0.227		-0.132	$-2.517 \cdot 10^{-4}$		$0.847 \cdot 10^{-4}$	$1.357 \cdot 10^{-4}$	0.876
LCa	4.626	-0.157	-0.399	-0.348	0.046				-0.110		0.045	0.045	0.773
$LSO_4{}^{2-}$ (p2, 3, 4)	3.340		-0.048	-0.069	-0.057		0.096	-0.099	-0.086				0.799
LMg (p1)	1.474								-0.109				0.832
LMg (p2, 3, 4)	2.095		-0.076	-0.076					-0.159				0.918
LK (p1)	0.464							-0.310	-0.070				0.579
LK (p2, 3, 4)	1.703				-0.216		-0.133	-0.376	-0.155				0.764
LCl (p1, 2)	2.595	0.173			-0.114			-0.263	-0.036				0.765
LCl (p3, 4)	3.630				-0.332		-0.137	-0.428	-0.124			0.026	0.892
LNa	2.269	0.229	0.842	0.979	-0.200	-0.132	-0.132	-0.200	-0.051		-0.079	-0.079	0.897
LPO_4^{3-} react.P	2.860	0.868	0.868						-0.212			0.180	0.561

a_{p2}, a_{p3}, a_{p4}: modify coefficient a to take into account sampling point.
a_{sp}, a_{Sm}, a_{Aut}, a_{Wt}: modify coefficient a to take into account seasonal influence.
b_2, b_3, b_4: modify coefficient b to take into account sampling point.
r: multiple correlation coefficient.

autumn), has as a consequence an intensification of the flow coefficient in the lower points of the river as compared with the upper ones ($b_{p1,2} = -0.036$; $b_{p3,4} = -0.124$).

The nitrate, ammonia, reactive phosphorus and total phosphorus concentrations cannot be adjusted as well as the other variables to the mathematical models tried in this study. In the case of total phosphorus and ammonia-nitrogen the relationships between concentration and flow are not significant.

Nitrate (see Fig. 8) shows a positive relation with flow, with greatest concentrations at times of large flows (Casey and Clarke, 1979; Edwards and Brooker, 1984). The nitrate ion is a chemical species with great mobility in the soil (Tisdale *et al.*, 1985), being easily removed by the rainfall percolating through the soil. This process could explain the increase in concentrations as flow increases (Casey and Newton, 1973). In contrast, ammonia, which has lower mobility in the soil, is not so positively correlated with flow.

Total phosphorus behaviour is very similar to ammonia (see Fig. 9). While the reactive phosphorus is correlated with flow, although not very strongly, total phosphorus does not show any pattern of behaviour. The increase in concentrations is possibly due to fertiliser applications to the farm land with leaching of nutrients

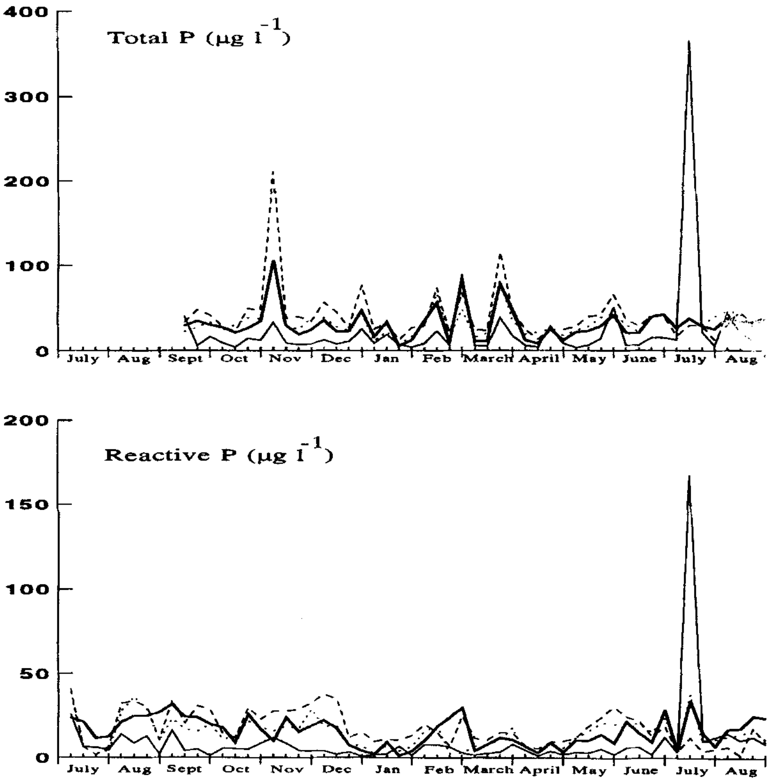

Figure 9 Variation of total and reactive phosphorus (μg l^{-1}) for the different sampling points. (—— Guerricaiz; ▬▬ Aulestia; ...... Guizaburuaga; _ _ _ Oleta).

after heavy rain. Corresponding increases in potassium concentration support this theory (Westlake *et al.*, 1972). These changes influence the degree of adjustment of the models obtained. The increase in phosphorus (365.6 μg Total-P l^{-1}, 167.4 μg Reactive-P l^{-1}), ammonia (100 μg NH_3-N l^{-1}) and potassium (4.59 mg K l^{-1}), observed in July 1986 in the samples from Guerricaiz, could be related to river pollution caused by a late application of fertilizer to pastures in the summer in order to obtain a second cut of grass.

CONCLUSIONS

The Lea River is a relatively unpolluted river, in which biological processes and the geological nature of the terrain are the main factors explaining the chemical composition of the waters.

Seasonal fluctuations in conductivity, alkalinity, magnesium, calcium, sodium, potassium, chloride and sulphate are explained basically by the flow variation and account for 59% to 90% of the variability.

Spatial variations, except for nitrate, are significant for all the chemical variables studied, and are mostly due to geological features of the catchment. The geology divides the catchment in two well marked areas: an upper catchment up to Aulestia, and a lower catchment to the river mouth. In the lower catchment an increase of sodium and chloride levels is observed. The marine influence is suggested as a cause for this phenomenon.

The levels of nitrate, ammonia, reactive phosphorus, total phosphorus and potassium show large increases at times of heavy rain; these increases might be due to wash-out of these elements applied as fertilizers to the surrounding land.

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