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A neuro-fuzzy modeling tool to estimate fluvial nutrient loads in watersheds under time-varying human impact

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Abstract

Fluvial nutrient loads are usually calculated through a bilogarithmic regression relating flow and river nutrient concentration. This relationship, however, can be highly nonlinear, due to changes in watershed land uses over time. Also, retransformation of data can result in important biases, and available databases usually do not provide the statistical properties needed to apply parametric statistics or time-series analysis methods. The validity and advantages over customary methods of an Adaptive Neuro-Fuzzy Inference System (ANFIS) for estimating fluvial nutrient loads in watersheds under time-varying human impact was tested. Fluvial nutrient loads time-series were modeled in two watersheds of different size and human impact history. ANFIS and methods based on rating curves and ratio estimators were applied to compare results. The ANFIS approximation gave unbiased estimates of loads and showed advantages over the other methods: It allowed the implementation of a homogeneous, model-free methodology throughout the data series, avoiding the presence of artifacts in the final load histories; it fitted the observed concentration time-series better than the other procedures; it worked in real space without the need to logarithmically transform and retransform data; and it gave annual dispersion values, which could be interpreted as annual uncertainties. In addition, the parameters fitted during the ANFIS modeling could be ecologically interpreted, and were a valuable tool to describe features of modeled data and to understand historical changes in human impact on watersheds. MATLAB codes and instructions to implement the new method are provided.

Cultural eutrophication assessment and control are amongst the most important issues natural resource managers must face, markedly in watersheds where agricultural practices involve intensive use of fertilizers (Halweil 2002). Excess of man-induced nutrient loading into rivers has not only driven freshwater eutrophication (Vollenweider 1968; Heaney et al. 1992; Reynolds 1992) but also degradation of coastal areas and resources at a global scale (Walsh 1991; Alexander et al. 2000; McIsaac et al. 2001). Despite other factors playing a role, diversion of nutrient inputs often leads to the improvement of water quality and the lessening of negative impacts of eutrophication (Reynolds 1992). Thus, the evaluation of river nutrient loads is of great importance in supporting water management decisions in polluted watersheds.

Frequently, it is possible to estimate the fluvial nutrient load to a water body over periods of several years or decades, allowing not only the definition of realistic target situations for managers, but also a better understanding of the effects of human activities over the watershed. In most situations, such load estimates (i.e., the sum of the product of constituent concentration and flow, over a given period) are calculated from high frequency river flow data (i.e., daily), whereas river nutrient concentration data are generally scarce, weekly at best. Several methods to calculate nutrient loads have been proposed to deal with such datasets (*see* Cohn [1995] for a review). Amongst the most popular are the ratio estimators (REs) and the rating curves (RCs). REs, based on the sampling theory (Cochran 1977), assume a constant ratio between constituent concentration and flow. If certain statistical assumptions are fulfilled, they perform well in a broad range of hydrological situations, basin sizes, and flow versus nutrient concentration

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relationships. They can also work with low nutrient sampling resolution. For details and examples, see Dolan et al. (1981), Richards and Holloway (1987), Webb et al. (1997), Preston et al. (1989), and Muckhopadhyay and Smith (2000). On the other hand, RCs build a continuous nutrient concentration trace (i.e., daily) from an empirical log-log relationship between available data on nutrient concentration and flow. Indications for their correct application and various modifications on the original method are found elsewhere (Dolan et al. 1981; Ferguson 1986, 1987; Cohn et al. 1989; Clarke 1990; Gilroy et al. 1990; Cohn et al. 1992; Cohn 1995; Lennox et al. 1997; Webb et al. 1997, 2000; Phillips et al. 1999; Robertson and Roerish 1999; Clement 2001).

When calculating loads for long periods (i.e., years or decades), the assumption of a constant nutrient concentration versus flow relationship assumed by RC and RE methods is not always realistic, due to the presence of nonlinear dynamics in the relationship. This is particularly true in watersheds of developed countries, where land and water uses have experienced dramatic changes during the last decades (see CENR [2000] for an example). Different solutions have been proposed to calculate loads in such situations. The most simple and widely applied solution is to use the methods listed above on portions of the dataset with homogeneous nutrient concentration versus flow relationship (Preston et al. 1989; Clement 2001). This technique, however, has several drawbacks: it needs subjective decisions when grouping the years, and inference in the limits of each relationship is uncertain, usually resulting in unrealistic steps in the final load time-series. Van Dijk et al. (1996) proposed a moving-window approximation, in which regression models were successively fitted to data grouping years in a moving-average fashion. However, estimates for the final period of calculation tend to be significantly biased (Stålnacke and Grimvall 2001). Cohn et al. (1992) used multivariate regression to account for time trends and seasonality. Limitations of their approach will be outlined later in this paper. Stålnacke and Grimvall (2001) used a refinement of the application of several linear regressions, but their approach only accounts for linear relationships, and the risk of overfit is not clearly managed. Other authors (Hipel 1985; Vecchia 1985; Zetterqvist 1991) proposed using time series analysis (e.g., transfer functions and autoregressive models). Despite the robustness of some of these applications, using time series analysis on a given series is a difficult and time-consuming task, because the number of possible models is very large. Also, a high degree of expertise is needed to apply these analyses (Legendre and Legendre 1998), and available data are not always suitable for working with these approaches, mainly because the sampling time step is seldom constant. Thus, the applicability of time-series analysis methods is very restricted.

The aim of this paper is to present neuro-fuzzy modeling as an alternative tool for calculating long-term fluvial loads in watersheds where flow versus nutrient concentration relationships are strongly nonlinear and uncertain due to the human impact history. The application of neural networks to modeling nonlinear relationships in aquatic sciences has grown rapidly during the last three decades (Lek et al. 1996; Lek and Guégan 1999; Maier and Dandy 2000). However, one of its main disadvantages is that neural networks act as black-box inference machines. Fuzzy logic sets, on the other hand, are based on transparent, editable linguistic rules, and establish a practical framework to include human expertise into modelization (Borri et al. 1998). In this paper, we merged the fuzzy logic approach with the ability of learning algorithms from neural networks to empirically adjust a model to an input-output problem. We applied this approach, and the RC and RE methods, to calculate historical nutrient loads in two watersheds under strong human impact, to show advantages of the neuro-fuzzy approach over the other methods.

Neural networks and fuzzy logic methods are little used in limnology, and almost completely ignored in classical statistics textbooks. Moreover, standard statistical packages do not implement these procedures. To avoid these limitations, and to offer an open technique (avoiding ad hoc solutions that are difficult to implement in different scenarios), we provide a MATLAB application to calculate annual loads using the method presented here. Recommendations are also given for its correct use.

Materials and procedures

Materials—We modeled two long-term river nutrient concentration series in two watersheds with environmental problems related to human activities, i.e., eutrophication of Sau Reservoir in the watershed of Ter River (Spain) and Gulf of Mexico hypoxia in the mouth of the Mississippi River (U.S.A.).

Ter River watershed: Eutrophication of Sau Reservoir—Sau Reservoir was first filled in 1963 in a middle stretch of the Ter River (Fig. 1A), as part of a multi-use scheme, including hydroelectric power, agricultural irrigation, domestic and industrial water supply to metropolitan areas, and recreational activities. Sau Reservoir drainage basin is a 1380 km^2 populated area (109 people km^{-2}), mainly covered by woodland (78%) and agricultural land (16%). Ter River median flow is 10 $m^3\ s^{-1}$. Since the Sau Reservoir was built, the water body experienced a process of increasing eutrophication (Vidal and Om 1993), from moderately eutrophic during the first years to severe eutrophication in the late 1980s. Several human activities in the basin contributed to the process: intensive use of fertilizers, pig and stock farming development, proliferation of industrial areas, and changes in land use (Sabater et al. 1990; Sabater et al. 1991; Vidal and Om 1993; Sabater et al. 1995; Espadaler et al. 1997). Wastewater treatment plants (WWTP) were built in main urban and industrial areas during the early 1990s (Vidal and Om 1993), leading to a moderate improvement of the water quality of the reservoir.

Limnological characterization of Sau Reservoir started in 1963, conducted by the local water supply company (Aigües Ter Llobregat). Thus, a good database is available on both

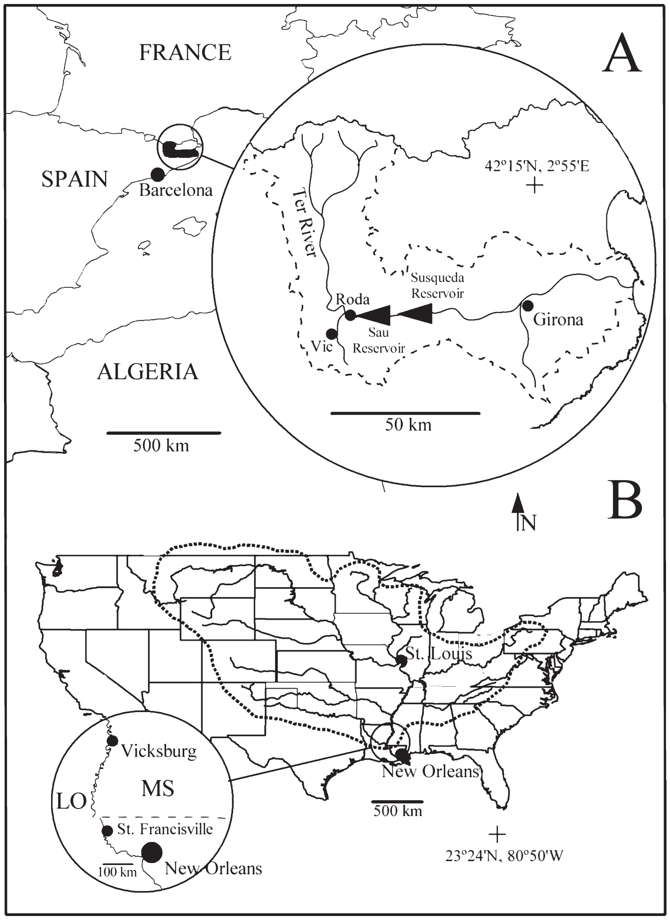

Fig. 1. (A) Location of the Ter River watershed and Sau Reservoir. (B) Location of the Mississippi River watershed and of sampling points.

changes in the water body and on incoming materials through the main inflow (the Ter River, which accounts for 85% of land drained by Sau Reservoir). Total phosphorus (TP) river concentrations were used to estimate phosphorus loads to the reservoir. Analysis for TP in Ter River at Roda de Ter (Fig. 1A) was initiated in 1972. Sampling was weekly to monthly, and no stratification upon flow or season was applied (i.e., the sampling frequency was independent of flow or season). There are three gaps in the series: year 1983 and year 1986, and the period 1991 to 1994 (both inclusive). These years were not included in the final load histories. A total of 414 samples were analyzed for TP concentration (Fig. 2A), using the alkaline persulfate oxidation method (Grasshoff et al. 1983).

The government water agency (Agència Catalana de l'Aigua) supplied the daily averaged flows for the period 1972 to 1997 (Fig. 2A) measured at a gage-station located 1 km upstream from the TP sampling point. From 1998, daily flows were estimated by the Agència Catalana de l'Aigua from water budgets of Sau Reservoir. No significant differences were found between gage-station estimates and water-budget estimates calculated for years prior to 1998.

Mississippi watershed: Hypoxia in the Gulf of Mexico—Since the early 1980s, much attention has been paid to the hypoxia forming in front of the coast of Louisiana, and many efforts have been focused on finding causes and solutions for the observed oxygen depletion (CENR 2000). Authors have reported a direct relationship between nitrate load from the Mississippi River, which has increased over the last decades, and hypoxia development in the Gulf of Mexico (Goolsby et al. 1999; CENR 2000; Goolsby and Battaglin 2001). The Mississippi watershed is 3,208,700 km^2, with a population density of 22 people km^{-2}. Main land covers are agricultural land (58%), barren land (21%), and woodland (18%). Median flow in the Mississippi River is 14,896 m^3 s^{-1}.

We estimated nitrate load in the lower Mississippi River for the period 1955 to 1997. River nitrate concentration data at St. Francisville (Fig. 1B) were obtained from the United States Geological Survey (USGS) National Water Information System, a public-domain computerized database (http://waterdata.usgs.gov/nwis, USGS Station Number for St. Francisville: 07373420). Sampling for nitrate was 6 to 15 times per year. Total number of samples was 753 (Fig. 2B). Nitrate was analyzed by the phenoldisulfonic acid method (Rainwater and Thatcher 1960) prior to the early 1970s, and by an automated cadmium reduction method afterward. Goolsby and Battaglin (2001) confirmed that the change in method had no systematic effect on the data, and data for the whole period were treated together. Mean daily streamflow data at Vicksburg (Mississippi) for the period 1955 to 1997 (Fig. 2B) were obtained from the USGS database (USGS Station Number for Vicksburg: 07289000).

Procedures—We applied three different approaches to nutrient load calculations in the two modeled basins, in order to compare advantages and limitations of classical procedures with the new method presented here. Loads were estimated with software that implements classical RC and RE methods with a modified multivariate RC accounting for time trends, and with the proposed neuro-fuzzy inference system.

FLUX software—The FLUX software was developed by the US Army Corps of Engineers (Walker 1996), as part of a package written to bring procedures for eutrophication assessment and prediction. FLUX is available at no cost at http://www.wes.army.mil. The program is used to estimate nutrient loads, or other water quality components, going through a tributary sampling station over a given period. Using six calculation techniques (Table 2.1 in Walker 1996), FLUX applies the flow versus nutrient concentration relationship developed from the sample record onto the entire flow record to calculate total mass discharge and associated error statistics, calculated by a jackknife procedure (Walker 1996). The program includes the option of stratifying the data, i.e., grouping the data upon flow and/or season. It also offers a variety of graphic and tabular output formats to assist the user in evaluating data adequacy, and in selecting the most appropriate calculation method and stratification scheme for each application.

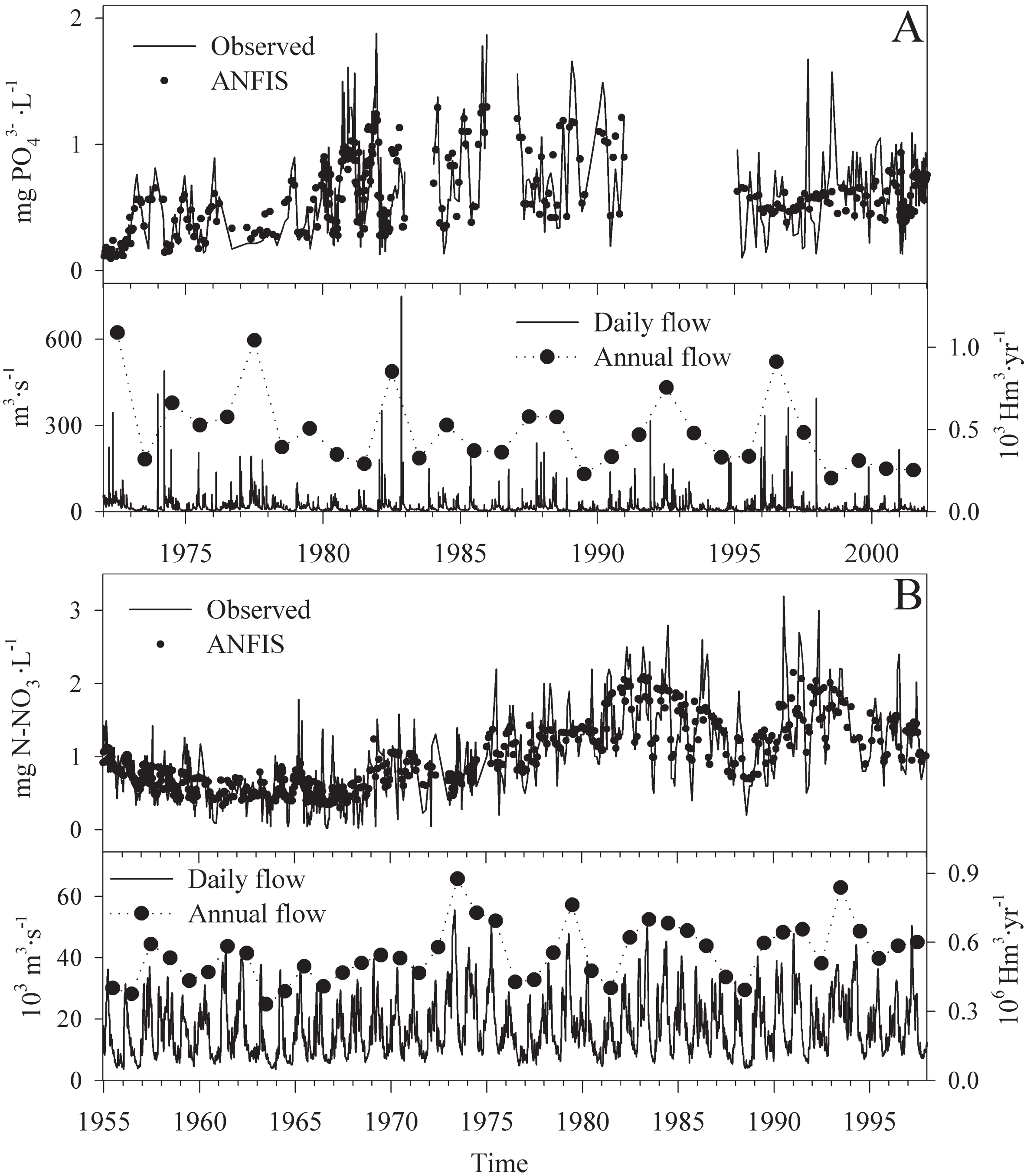

Fig. 2. Observed time-series of river nutrient concentration and flow in (A) the Ter River and (B) the Mississippi River. The top figure in each panel shows the observed river nutrient concentration and the corresponding ANFIS reconstructed time-series. Each dot is the mean value for 1000 iterations. The bottom figure in each panel shows the daily mean flow (left axis) and the annual flow (right axis) for the studied period. Ticks in time axes represent January 1.

The procedure to calculate nutrient loads in the two systems under study was as follows:

1. The series was split into segments considered homogeneous regarding the nutrient concentration versus flow relationship (a bilogarithmic linear regression was applied). We calculated loads for groups of consecutive years if no significant time trend was present in the residuals.
2. The optimal calculation method and stratification scheme was then applied to each group of data, while considering multiple criteria, i.e., suitability of the method to data,

Table 1. FLUX methods implemented during simulation of each period considered in the series of Ter and Mississippi rivers

Period grouped for calculation	Number of samples	Load estimation method*	Stratification scheme†	Standard deviation of load estimation‡
Ter River				
1972	23	Rating curve (5)	2 strata by flow	4.50
1973-1979	68	Rating curve (5)	2 strata by flow	4.40
1980-1984	120	Ratio estimator (2)	3 strata by flow/season	6.60
1985-1990	49	Ratio estimator (3)	3 strata by flow	5.80
1995-2001	155	Ratio estimator (2)	3 strata by flow	8.20
Mississippi River				
1955-1956	72	Ratio estimator (2)	3 strata by flow/season	3.50
1957-1960	117	Ratio estimator (2)	3 strata by flow/season	3.10
1961-1974	302	Ratio estimator (2)	3 strata by flow	3.20
1975-1977	36	Ratio estimator (2)	2 strata by flow	5.30
1978-1980	36	Ratio estimator (2)	2 strata by flow	4.70
1981-1986	70	Ratio estimator (2)	3 strata by flow/season	3.20
1987-1989	34	Rating curve (5)	2 strata by flow	5.00
1990-1993	44	Rating curve (5)	2 strata by flow	5.10
1994-1997	42	Ratio estimator (2)	3 strata by flow/season	4.00

*Numbers in brackets are method numbers in Table 2.1 in Walker (1996).
†The load estimation method is applied independently to each stratum. Flow strata are defined by a built-in procedure in FLUX (Walker 1996), and seasonal stratification is optimized by residual plots analysis.
‡Calculated by a jackknife procedure (Walker 1996) and expressed as percentage of estimated load.

residual analysis, and standard deviation in load calculations (for details on decision making, *see* Walker 1996).

Table 1 shows methods and stratification schemes used in each segment defined by the FLUX procedure in the Ter and Mississippi rivers.

Multivariate rating curve—A single multivariate model may be used to calculate series with time-varying nutrient concentration versus flow relationships, instead of the classical multiple RCs and REs. Cohn et al. (1992) proposed a multivariate rating curve to account for nutrient concentration versus flow relationships and time trends and seasonality in nutrient concentration, i.e.,

$$\ln[C] = \beta_0 + \beta_1 \cdot \ln\left[\frac{Q}{Q'}\right] + \beta_2 \cdot \left\{\ln\left[\frac{Q}{Q'}\right]\right\}^2 + \beta_3 \cdot [T - T'] + \beta_4 \cdot [T - T']^2 + \beta_5 \cdot \sin[2 \cdot \pi \cdot T] + \beta_6 \cdot \cos[2 \cdot \pi \cdot T] + \varepsilon \quad (1)$$

where $\ln[C]$ is the natural logarithm of the constituent concentration C, Q is discharge, T is time (years), and ε is an error term. Q' and T' are centering variables that set the terms of the polynomials orthogonal (Cohn et al. 1992). β_i is empirically estimated parameters. A minimum variance unbiased estimator was employed to correct for the retransformation bias (Cohn et al. 1989). Variances of annual loads were calculated as the sum of daily load variances plus the covariance terms (Likes 1980; Gilroy et al. 1990). Calculations were implemented in a standard statistical package (SAS System) and a spreadsheet (Microsoft Excel).

Adaptive neuro-fuzzy inference system—A fuzzy inference system (FIS) is a computing framework that combines the concepts of fuzzy logic, fuzzy decision rules, and fuzzy reasoning (Jang 1993; Jang and Sun 1995).

The fuzzy logic theory was first formulated by Zadeh (1965) as a new way of characterizing nonprobabilistic uncertainties. In contrast to the Boolean 1-0 logic, fuzzy logic also permits in-between values for any judged statement, i.e., it applies a continuous, multi-valued logic between 0 and 1. In terms of a function relating a variable with the probability associated to a judged statement (membership function [MF] hereafter), this is equivalent to replace a rectangular MF with a smoothed MF (*see* the Numerical Example file in Appendix A). Fig. 3 gives examples of Gaussian MFs, a common choice in fuzzy logic applications.

The fuzzy decision rules are the way an FIS relates an input variable (say X) to an output variable (say Z). They take the form:

$$\text{if } x \text{ is A, then } z \text{ is B} \quad (2)$$

where A and B are linguistic values (low or high, for instance) defined as MF in the input and output spaces. We can employ more than one variable in the premise side and construct rules such as

$$\text{if } x \text{ is A and } y \text{ is B, then } z \text{ is C.} \quad (3)$$

Fuzzy reasoning is an inference procedure used to derive conclusions from a set of fuzzy decision rules. The steps of fuzzy reasoning performed by an FIS are (Jang 1993):

1. Compare the input variables with the MFs on the premise part of the fuzzy rules to obtain the probability of each linguistic label (fuzzification).

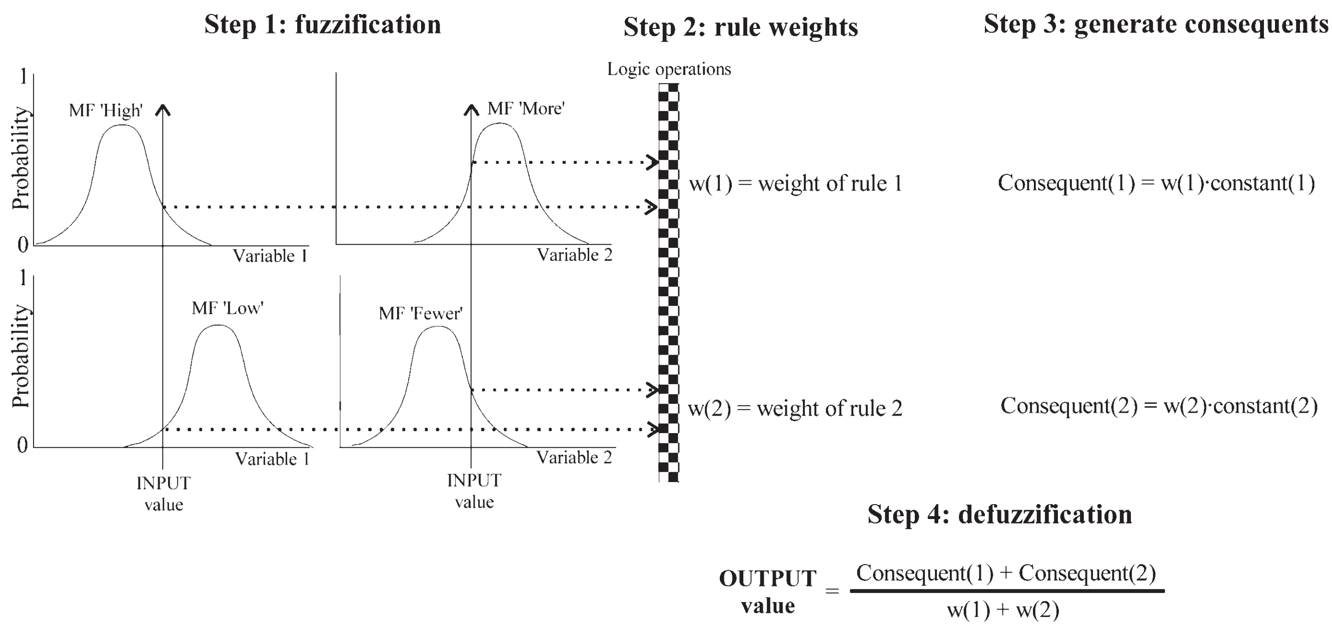

Fig. 3. Steps during fuzzy reasoning. The example consists in a zero-order Sugeno type FIS (*see* text for details), with two inputs, one output, and two if-and-then rules. Each input space has been characterized by two intuitively labeled Gaussian MF, drawn separately for clarity and to give graphical representation of each rule. The reader should start reasoning from assigned values to inputs, and then follow arrows and explanations in the text.

2. Combine (through logic operators) the probability on the premise part to get the weight of each rule. For a comprehensive treatment of the use of logic operators in fuzzy logic, *see* Jang and Sun (1995).
3. Generate the qualified consequent of each rule depending on their weight.
4. Aggregate the qualified consequents to produce a crisp output (defuzzification).

Fig. 3 gives a graphical example of fuzzy reasoning. In this case, the MF of the consequent of each rule is a constant instead of a fuzzy MF. These kinds of FIS are called zero-order Sugeno-type and are very convenient for fitting procedures (Jang et al. 1995). The Numerical Example file in Appendix A contains a detailed example of fuzzy reasoning.

Given an input-output problem, the construction of an FIS has two fundamental steps: the specification of an appropriate number and type (Gaussian, triangular, and so on) of input and output MFs (structure identification), and the specification of the shape of the MFs (parameter estimation). Whereas the structure identification is solved by human expertise or trial-and-error, numerical methods have been proposed to solve the parameter estimation step. Wang and Mendel (1992) reported a method based on the generation of fuzzy rules by examples from the modeled data set merged with human expertise. Buzás (2001) successfully applied this methodology to nutrient load estimations in tributaries to the Lake Balaton. In this paper, we used a different approach, which takes advantage of adaptive neural networks algorithms during fitting procedures. As stated by Jang (1993), an Adaptive Neuro-Fuzzy Inference System (ANFIS) is a Sugeno-type FIS where the MF parameters are fitted to a dataset through a hybrid-learning algorithm.

ANFIS implementation—The core of the ANFIS calculations was implemented in a MATLAB environment. Functions from the Mathwork's MATLAB Fuzzy Logic Toolbox (FLT) were included in a MATLAB code programmed by the authors, to apply the ANFIS load estimation in a Monte-Carlo framework, and to obtain diagnostic analyses. The code to implement ANFIS, a step-by-step manual, and recommendations for potential users are in Appendix A.

For load calculations with ANFIS, the same problem was considered for the two scenarios. We defined the mean daily flow and time (expressed as number of days since January 1 of the first year in the dataset) as inputs. The output was the constituent concentration (TP for Ter River and nitrate for Mississippi River).

The structure identification was solved applying a trial-and-error procedure and a conservative criterion (i.e., minimum number of parameters to the best fit). Consistent with this, Gaussian MFs were used in the input spaces, as only two parameters suffice to define this curve. The ANFIS function of the MATLAB FLT was used to solve the input-output problem above with different numbers of input MFs, using all data available. An estimate of the mean square error between observed and modeled values were computed for each trial, and the best structure was determined considering a trade-off between the mean square error and the number of parameters involved in computation. Input MFs were linked by all possible combinations of if-and-then rules (Eq. 3), defining an out-

Table 2. Performance of the tested methods predicting daily river nutrient concentrations in the Ter and Mississippi rivers*

Method	Explained variance (r^2)	Residual trend analysis: Residuals vs. time†	Residuals vs. flow†	Residuals vs. predicted values†	Test for normality‡
Ter River					
ANFIS§	0.60	0.18	0.38	0.49	<0.01
FLUX	0.52	<0.05	<0.001	0.31	<0.001
Cohn	0.51	0.34	0.16	0.31	<0.001
Mississipi River					
ANFIS§	0.63	0.14	0.39	0.15	<0.001
FLUX	0.58	0.21	<0.01	0.43	<0.05
Cohn	0.33	<0.001	<0.001	<0.001	<0.01

*The whole period is considered in each case.
†P value of the Kendall rank coefficient test for presence of trends in the residuals. H_0 = no trend.
‡P value of the Kolmogorov-Smirnov test for normality of the residuals. H_0 = normality.
§Average nutrient concentration for the 1000 ANFIS time-series generated (Fig. 2).

put constant for each rule. Details on this procedure are given in the User Manual and Numerical Example files in Appendix A. In the Ter River case, this procedure gave 6 MFs in each input (36 rules and output constants), whereas in the Mississippi River gave 15 MFs in the 'Time' input and 4 MFs in the 'Flow' input (60 rules and output constants).

Once the FIS structure was identified, the parameters that had to be estimated (Gaussian input MF parameters and output constants) were fitted by the hybrid-learning algorithm (HLA hereafter) implemented in the ANFIS FLT function. To avoid overfitting problems during the estimation, the data set were randomly split into two sets: a training set (2/3 of the data), used by the HLA to fit, and a checking set (1/3 of the data), which was not used by the HLA. When both checking data and training data were presented to ANFIS, the FIS was selected to have parameters associated with the minimum checking data model error.

There is no analytical expression to calculate variances of loads obtained through ANFIS. To solve this limitation, and also to avoid biases that could arise by using only one random sample, the inference procedure explained above was implemented in a Monte Carlo sampling framework. The database was randomly resampled without replacement 1000 times, maintaining the ratio between training and checking sets. Thus, 1000 daily concentration series were generated, and consequently, we obtained 1000 annual load estimates for each year. The unbiased annual load for each year was the average of the corresponding 1000 loads, and precision values were calculated as the variance of associated distributions. Ten of the 24 collections of 1000 annual load estimates in the Ter River scenario showed non-normality (Kolmogorov-Smirnov test P value < 0.05). However, the small deviation of the median respect to the average (2.4%, n = 10) suggested that the average was an unbiased parameter of the central tendency, and indices of bimodality were present only in years 1977 and 2000. In the Mississippi River calculations, six of the 43 collections of 1000 annual load estimates showed non-normality (Kolmogorov-Smirnov test P value < 0.05), but the small deviation of the median respect to the average (0.6%, n = 6) also suggested the average was an unbiased parameter of the mean, and no bimodal distribution was observed.

The performance of ANFIS estimation mainly depends on the completeness of the database (Costa Branco and Dente 2001). However, when dealing with nutrient load calculations, this prerequisite is difficult to achieve, as samples for nutrient analysis are usually comparatively scarce. In such situations, information holes in the fuzzy model could appear, resulting in nonsense values in the modeled series (i.e., negative nutrient concentrations or values several orders of magnitude above the maximum concentration found in the river). These nonsense values represented an average of 1.8% of the modeled values from data of the Ter River (nonsense values from gaps included), and an average of 0.99% from data of the Mississippi River. Nonsense values were replaced by the nutrient concentration of the preceding day in the modeled series.

Assessment

The assessment of the new method for estimating fluvial nutrient loads was divided into four steps: (1) a comparison between the nutrient concentration time-series obtained with the three procedures. All methods to calculate nutrient loads directly (ANFIS, RCs) or indirectly (REs) use a predicted daily nutrient concentration time-series. Despite the fact that nutrient concentrations are not the target of these methods, this underlying calculation is of great importance, because a poor performance predicting nutrient concentrations will lead to a poor performance predicting loads. (2) A comparison between the nutrient load time-series predicted by the three methods. (3) A performance statistical analysis of the ANFIS approach, as a nonrelative way to assess the ability of ANFIS calculating loads; and (4) an examination of the ability of the fuzzy modeling transparent rule feature to explain changes in the nutrient concentration versus flow relationship, because straightforward interpretation of estimated parameters is considered one of the main features of the new technique.

Nutrient concentration time-series comparison—Table 2 summarizes the results obtained comparing the observed nutrient concentration time-series (Fig. 2) with the time-series calculated by the three load calculation methods. The ANFIS time-series (Fig. 2) was the result of averaging the 1000 daily time-series obtained in each case. As shown in Table 2, the method that best fitted the observed data was ANFIS.

We also carried out residual trend analyses to detect systematic departures from observed time-series. The Kendall rank coefficient nonparametric test (Sokal and Rolf 1995) was used to detect trends in the residuals, because these showed significant departure from normality (Table 2) (Peters et al. 1997). ANFIS did not show any significant trend in the residuals. By contrast, both FLUX and Cohn methods showed significant systematic departures. It should be stressed that the application of FLUX method avoided trends in the residuals within each calculation group (Table 1), but this did not avoid systematic departures when looking at the whole period (Table 2).

Although Cohn's multivariate rating curve was statistically significant in both cases ($P < 0.001$), the low explained variance and results from residual analyses for the Mississippi River case suggested model misspecification. The plotted time-series of nutrient concentrations sampled from the Ter River (Fig. 2A) described a parabolic-like curve, which could be well fitted using a quadratic polynomial such as Eq. 1. The complex time-series sampled from the Mississippi River (Fig. 2B) could not be modeled by a quadratic fit, leading to spurious predictions. Despite results of the diagnosis analysis, we applied the model over the entire series (i.e., without splitting it), to illustrate the effect on final results of the highly nonlinear nutrient concentration versus flow relationship in this system. Thus, poor performance of the method reported in this case should not be considered as negative criticism of the method itself, but as limiting its applicability.

Nutrient load time-series comparison—Fig. 4 shows the nutrient load time-series in Ter and Mississippi rivers, calculated by the three methods under comparison, expressed as flow-normalized annual nutrient load and standard deviation. To compute flow-normalized loads:

$$Flow\,normalized\,load_i = \frac{Actual\,load_i}{Q_i} \cdot Q' \qquad (4)$$

where *actual load*$_i$ is the predicted load in year i, Q_i is the annual flow in year i, and Q' is the mean annual flow for the whole period. Flow-normalization eliminates the effect of the varying hydrology in the time-series plot. This makes the figure more readable, it allows the interpretation of the changes in the flow-normalized loads as indications of changes in the man-induced loading, and it better reflects the underlying nutrient concentration versus flow relationship (Stålnacke et al. 1999; Stålnacke and Grimvall 2001).

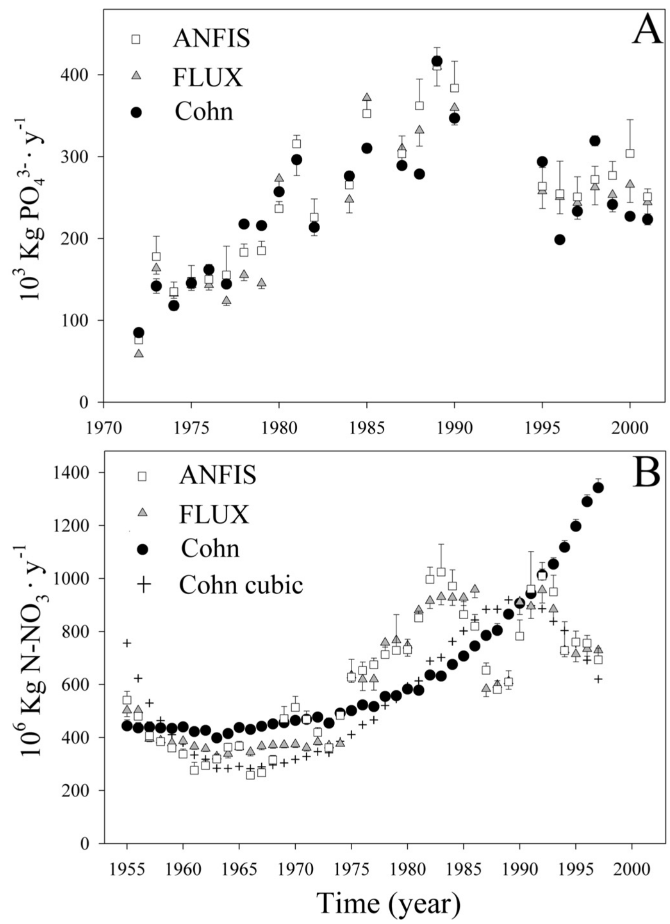

Fig. 4. Flow-normalized annual nutrient loads and standard deviation calculated through ANFIS, FLUX, and Cohn's procedures. (A) Results for the Ter River and (B) the Mississippi River. Results applying a cubic modification of Cohn's approximation are also shown in panel B. For clarity, only one error bar is represented for each symbol. Many error bars are unappreciable.

As shown in Fig. 4, results regarding general patterns of the series differ in the two rivers. Whereas in the Ter River the three methods gave similar results, (i.e., increasing nutrient load from 1972 to 1990, and decreases afterward), the Cohn's approximation gave dramatically different results than the other two methods used for Mississippi River calculations. This result is a direct consequence of the poor performance of Cohn's method predicting nutrient concentrations in the Mississippi River (Table 2). Cohn's model may be applied in such situations, splitting the series into fragments that can be quadratically fitted (*see* Goolsby and Battaglin 2001), procedure equivalent to that applied in the FLUX software. Increasing the degree of the polynomial in Cohn's equation did not give acceptable results (*see* Fig. 4B and Appendix B for derivation). Moreover, processing time and mathematical derivations increase formidably with the degree of the polynomial. Therefore, although Cohn's approximation could work in some cases, its applicability is limited.

ANFIS and FLUX calculations were both good in both scenarios. Detailed analysis, however, revealed the well-known step artifact in the FLUX series. Each transition between years modeled in different calculation groups (*see* Table 1) corresponds with a step in the final series. The artifact occurs in both plots, but is more evident in the Mississippi River FLUX series (*see*, for example, transition between years 1974 and 1975, 1977 and 1978, or 1986 and 1987). A step in the series could correspond with a real change in the river (in fact, some of the steps are followed by the ANFIS calculation), but the absolute correspondence between steps and transitions give strong evidence of presence of artifacts. By contrast, ANFIS applies a single method through the entire series avoiding the presence of artifacts (i.e., steps) in the final load histories. This adaptability to different situations and empirical relationships is possible due to two important features of the ANFIS approximation: (1) it is a model-free methodology, and (2) it acts as a local inference machine.

On the other hand, the FLUX and Cohn's approximation give averaged dispersion values, as the models are fitted to a group of years, or the entire series. Thus, interpretation of the standard deviation of individual annual loads is not straightforward. In contrast, the ANFIS method gives dispersion values independently calculated for each year. Thus, we can interpret standard deviations as annual uncertainties. It should be stressed that differences in annual uncertainty could arise from both the varying relationship between flow and nutrient concentration, and uncertainty caused by the different number of samples available for each year. Although no consistent relationship was found between number of samples and ANFIS calculated uncertainty in this study, this could not be the case in other situations. Thus, interpretation of ANFIS dispersion values as uncertainty in the flow versus nutrient concentration relationship should always take into consideration the effect of the sampling frequency. In any case, the possibility of analyzing these features of the data with annual independent calculated values is a great advantage of ANFIS over other procedures.

Performance of the ANFIS estimation—The validity of load estimations based on FLUX methods and the multivariate rating curve by Cohn et al. (1992) have been tested and confirmed on numerous occasions (Dolan et al. 1981; Richards and Holloway 1987; Preston et al. 1989; Clarke 1990; Cohn et al. 1992; Webb et al. 1997; Phillips et al. 1999; Muckhopadhyay and Smith 2000). To test the performance of ANFIS in calculating loads, we applied a split-sampling approach similar to that used by Preston et al. (1989) and Cohn et al. (1992).

The ANFIS method was tested separately at both sampling sites. The complete records of daily flow, time, and nutrient concentrations were randomly subsampled under different frequencies to obtain training and checking sets of different sizes (training set was 2/3 the sampled set; checking set 1/3 the sampled set). Data not included in the preceding sets constituted the evaluation set. The two first sets were used to train the ANFIS, using the model structures presented in subsection *Procedures*. Then, input data in the evaluation set was used to model a sum of loads with the FIS that were compared to the true sum of loads contained in the output of the evaluation set. Subsamples of the training + checking sets were of 50 samples, and calculations were repeated with 50 more samples in these two sets until the maximum number of samples in the original data set was reached. One hundred different calculations were done for each frequency. Nonsense values, as a result of inference from nontrained inputs, were eliminated and not replaced. They were registered and not used in final calculations.

Table 3. Performance of ANFIS estimation in Ter and Mississippi river scenarios (*see* text for details)*

Samples in training + checking sets	Mean percentage bias (SD)	Samples in evaluation set	Mean percentage of nonsense values (SD)
Ter River			
50	23.46*** (16.87)	364	25.8 (11.30)
100	10.90*** (13.30)	314	7.90 (4.45)
150	11.20*** (9.76)	264	4.90 (2.60)
200	6.00*** (10.10)	214	3.40 (2.47)
250	4.80** (9.65)	164	2.50 (1.77)
300	3.08ns (10.20)	114	1.90 (1.50)
350	4.42ns (11.50)	64	2.06 (1.53)
400	3.09ns (28.00)	14	1.57 (3.00)
Mississippi River			
50	–7.96ns (29.1)	703	31.63 (1.68)
100	–1.22ns (22.2)	653	26.29 (1.79)
150	–0.53ns (16.35)	603	15.42 (1.44)
200	–1.29ns (13.10)	553	8.89 (1.32)
250	–1.11ns (9.28)	503	5.72 (1.02)
300	–0.67ns (7.00)	453	4.04 (0.92)
350	–1.15ns (6.99)	403	3.37 (0.94)
400	–1.31ns (6.57)	353	2.67 (0.95)
450	–1.50ns (7.99)	303	2.25 (0.92)
500	–0.88ns (8.07)	253	1.81 (0.82)
550	–1.01ns (7.96)	203	1.56 (0.77)
600	–1.81ns (10.95)	153	1.48 (0.96)
650	–1.96ns (13.46)	103	1.41 (1.19)
700	–4.15ns (20.89)	53	1.26 (1.65)

*Statistical difference between estimated and true load: ns = not significant; ** = significant at the 1% level; *** = significant at the 0.1% level.

The mean percentage bias between the sum of loads estimated and the true sum of loads was calculated as follows:

$$B_x = \frac{\sum_{i=1}^{100}\left[100 \cdot \left(\frac{L_{e,i} - L_{r,i}}{L_{r,i}}\right)\right]}{100} \quad (5)$$

where B_x is the mean percentage bias for a given sampling frequency x, $L_{e,i}$ the estimated sum of loads for the ith random sampling, and $L_{r,i}$ the true sum of loads for the ith random sampling. The statistical significance of the difference between the estimated sum of loads and the true sum of loads was tested with the Mann-Whitney U test (Sokal and Rolf 1995).

Table 3 shows the results obtained from the performance analysis of ANFIS estimation. In the Ter River scenario, the mean percentage bias gave nonsignificant values with 300 samples in the training + checking sets. Thus, we considered load calculations in the Ter River as unbiased (Fig. 4A, 414 samples). With fewer samples, calculations were significantly biased, markedly with fewer than 100 samples. The mean percentage of nonsense values during performance analysis for 400 samples (Table 3) coincided with the mean percentage of nonsense values reported during simulations (i.e., 1.8%).

In the Mississippi River, the ANFIS procedure worked well even with few samples in the training + checking sets, and the mean percentage bias showed nonsignificant values with 50 samples. Consequently, load calculations in the Mississippi River (Fig. 4B, 753 samples) were also considered as unbiased. The mean percentage of nonsense values during performance analysis for 700 samples was slightly higher than the percentage found during simulations (i.e., 0.99%). This is likely due to the effect of the reduced number of samples in the evaluation sets (Table 3) compared to the number of simulated days in the load calculations (i.e., 15,706 days).

Ecological interpretation of ANFIS parameters—A desirable property of any model applied to real data are that the parametric structure of the model resulting from fitting procedures has physical meaning, i.e., that the model does not work as a blind fitting machine. A remarkable feature of the ANFIS approximation is that we can interpret fitted parameters. Fig. 5 shows the effect of all input combinations (flow and time) on the outputs (nitrate and total phosphorus concentration) in Mississippi and Ter rivers. Panels in Fig. 5 could be interpreted as the plot of all defined if-and-then rules (see Eq. 3) in each system, but also as a plot of the evolution over time of the nutrient concentration in each flow category. Some rules were not included in the graph because standard deviation of the associated output parameters were very high and made the figure unreadable. This is a consequence of information holes that arise from incomplete databases. Values for output parameters should be interpreted in a relative mode, because final concentrations are generated after defuzzification procedures.

As shown in Fig. 5A, nutrient concentrations in the Ter River were higher at low flows, suggesting a dilution dynamics governed by point sources located near the sampling point. In fact, man-induced nutrient loading in the Ter River watershed is concentrated around the industrial and farming area of Vic (Vidal and Om 1993), and the main stream draining this area (Gurri River) discharges few kilometers upstream the sampling point (Fig. 1A). Differences between periods were more evident in the lowest flow region, with the highest phosphorus concentrations during the periods just before 1993, when WWTP were placed in the basin. Dilution dynamics was also clearer before 1993. Thus, we can hypothesize that it was a change in the relative importance of point sources as a consequence of the WWTP implementation. This hypothesis is also supported by results from observed flow versus observed nutrient concentration log-log relationships. Regression for the 1980 to 1990 period (n = 169, P < 0.001, r^2 = 0.48) gave an intercept of 0.28 mg PO_4 L^{-1} and a slope of –0.47. Regression for the period 1995 to 2001 (n = 155, P < 0.001, r^2 = 0.10) gave an intercept of 0.19 mg PO_4 L^{-1} and slope –0.22. Differences between both intercepts and slopes were significant (P < 0.001, Tsutakawa and Hewett 1978). Considering the value of the intercept as an indication of the point sources level and the slope as an indication of the importance of the dilution effect (Behrendt 1993; Stålnacke and Grimvall 2001), this analysis supports conclusions extracted from ANFIS parameters.

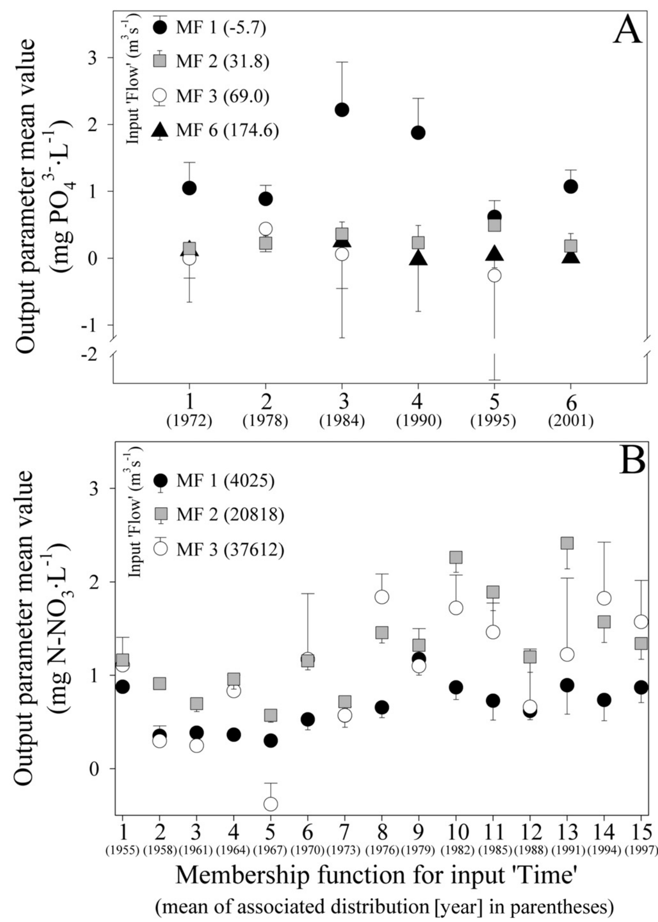

Fig. 5. Graphical representation of the if-and-then fuzzy rule structure obtained during nutrient load calculations in (A) the Ter River and (B) the Mississippi River. Symbols represent the mean value assigned by the HLA for each output constant parameter using 1000 iterations, and bars are standard deviations. (For clarity, symbols corresponding to rules with very high output parameter standard error were not drawn. Also, only one error bar is represented for each symbol. Many error bars are unappreciable). The average value of the mean parameter of each gaussian MF is given in brackets. Graphics must be read as follows: if MF for Flow is [symbol] and MF for Time is [x-value], then output constant is [y-value].

Error bars and missing rules inform about the uncertainty of the ANFIS inference. Uncertainty is present in the high flow regions, and poorly represented in the nutrient concentration sampling. This is a consequence of the hydrology of the Ter River (Fig. 2A), with stable low flow conditions and sudden floods typical from Mediterranean rivers. Uncertainty observed in the lowest flow region, represented well by the nutrient concentration sampling, was caused by the timing of point-sources spills, which could not be explained by flow. When WWTP were implemented in the basin the uncertainty was less pronounced, because most point sources were replaced by a single and more constant spill.

In the case of the Mississippi River (Fig. 5B), the uncertainty of the inference system also accumulated in the high flow regions. This is clearly shown by the standard deviation of the output parameter mean values and missing rules. This suggests that more sampling effort is needed during high flow periods. Nutrient concentration values were systematically higher at intermediate flows, suggesting nonpoint-sources prevalence (Behrendt 1993; Stålnacke and Grimvall 2001). This is also suggested by the observed flow versus observed nutrient concentration log-log relationship ($n = 714$, $P < 0.001$, $r^2 = 0.12$), which gave a positive slope of 0.29. In fact, runoff from agricultural land is thought to be the main nitrogen input to the Mississippi River (CENR 2000). Output parameter mean values followed an increasing trend (especially at intermediate flows), which is in agreement with the direct relationship established between the increasing amount of agricultural fertilizers applied in the basin and the nitrogen load transported by the river (Alexander et al. 2000; Goolsby and Battaglin 2001; McIsaac et al. 2001).

Discussion

Solutions available to calculate long-term loads in watersheds under time-varying human impact have several limitations. The most important are solved by the new method proposed here. The following is a list of the main advantages of the new technique:

1. ANFIS is a model-free, easy to implement approach. In contrast to time-series methods, little training is needed to calculate loads with ANFIS (*see* Appendix A for MATLAB codes and use). ANFIS implements a single-fitting procedure to complex (i.e., nonlinear) situations, without the need of establishing a formal model for the problem being resolved. Thus, no a priori information is needed about the empirical relationship between the explanatory and predicted variables, and the method suitability is always tested a posteriori.
2. With customary methods, variables frequently need transforming to enclose the problem into a linear relationship. Retransformation of results into real space is not straightforward and is dependent on the statistical properties of the constituent versus flow relationship. These properties do not always allow correct retransformation of variables, leading to significant biases (Cohn 1995). Time-series methods require a constant time step during sampling. Our method avoids these drawbacks.
3. ANFIS fits nutrient concentration time-series better than customary methods, with no systematic departure from observed data. This and the application of a single fitting procedure over the entire series avoid the presence artifacts (i.e., steps) in the final load series.
4. On the other hand, calculated dispersions with ANFIS can be interpreted as annual uncertainties. Load calculations are often the first step in eutrophication studies. The values generated during load calculations are then used to calibrate models, compare loads, and define target situations. If management decisions directly or indirectly depend on load calculations, then knowledge about uncertainty in load estimations becomes a priority (Reckhow and Chapra 1983, 1999). Using annual dispersion values calculated from a classical, empirical relationship grouping several years could be inaccurate, unless variance is constant all over the relationship, and we have equal number of samples for all years.
5. Also, the ANFIS method allows interpreting the values of the parameters fitted. The transparent rule structure of ANFIS allows the user to extract information about the empirical relationship between flow and concentration over time, drawing concise explanations. This a posteriori interpretation seems preferable to the a priori constriction of the data to an empirical relationship with an often dubious physical meaning. This open-box feature makes ANFIS an attractive exploratory data analysis tool, especially in situations were available models fail explaining observed phenomena.

Comments and recommendations

The ANFIS procedure presented here, despite being model-free, must be implemented with care. A critical number of samples are needed to have significant results and to avoid having too many nonsense values during simulation. The database must be as complete as possible, including nutrient concentration samples over a broad range of flows and times (in a classical framework, working with no-trained data would be equivalent to extrapolation). It is best to exclude large gaps in the database from final results. We do not recommend any minimum number of samples per year, because this will strongly depend on the dynamics of the system. Alternatively, we strongly recommend using the performance analysis procedure explained in the text (*see* also Appendix A), and the fit between observed and modeled nutrient concentration time-series as a definitive criterion to judge the suitability of the method. If percent bias given by performance analysis is higher than 5%, results should be regarded with caution, and biases higher than 10% should not be accepted. However, because ANFIS uses a learning algorithm, we must always work with a number of model parameters not exceeding 1/4 the number of samples in the training sets, in order to avoid the risk of overfitting and loose generality. This places, in practice, the minimum number of samples at around 150.

The performance of ANFIS calculating loads strongly depends on the relationship between flow and river nutrient concentration. It should be stressed that in some rivers nutrient dynamics is independent of flow (Whitfield 1982), and that presence of large lakes and impoundments significantly alter flow and nutrient dynamics (Kelly 2001). Also, we tested our method in two basins covering a wide range of sizes, but not in very small basins where nutrient dynamics could be more unpredictable (Bernal et al. 2002). In these situations, ANFIS performance using flow as a key input variable could be low. This poses the question on how strong the relationship between flow and nutrient concentration should be. Our experience is that using regression to explore this relationship in long-term data are not optimal, because the time-varying effects mask the relationship and usually lead to a confusing and noisy scatter graph. Our recommendation is using ANFIS method, which is not time consuming, as an exploratory data analysis tool, and to judge the suitability of the method on the basis of the performance analysis on nutrient load calculations. If bias in load calculations is unacceptable, the analysis of the parameters fitted and the modeled nutrient concentration time-series could give valuable information to apply alternative procedures.

As a concluding remark, we used ANFIS to solve nutrient load calculations, but this technique should also work with few modifications in a broad range of regression problems. We encourage researchers to apply and modify our codes to implement ANFIS in other situations. Also, the exploratory data analysis ability of this technique deserves more detailed studies.

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