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Aquatic Sciences

Long-term trends of physical, chemical and biological variables in the River Danube 1957–1995: A statistical approach

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ABSTRACT

A 39-year series (1957 – 1995) of data on fourteen physical, chemical and biological variables from the Austrian section of the River Danube west of Vienna – Nußdorf was analysed statistically to detect long-term trends of the variables in relation to human activities (represented by time), discharge and water temperature.

Principal component analysis distinguished four main components explaining 72 % of the total variance: PC1 contains total phosphorus, soluble reactive phosphorus, ammonium and potassium permanganate values, PC2 contains nitrate-N, chloride and oxygen, PC3 contains BOD5 and nitrite-N, and PC4 contains numbers of bacteria. Trends in time were most pronounced for variables in PC1, but also occurred in PC2; variables in PC3 and PC4 had no trends. Seasonal patterns were marked for variables in PC2, slightly less in PC1, and least in PC3 and PC4. Concentrations were minimal in summer and maximal in winter, inversely related to discharge (maximal in summer). Following reductions in point-source nutrient inputs to the Danube, in both Germany and Austria, mean concentrations in the river have fallen by at least half since the 1980s. Chloride and nitrate-N also show trends towards lower concentrations. The Danube is well-oxygenated, with concentrations near air saturation values.

A mathematical relationship between concentrations of the variables and river discharge (Q), water temperature (T) and time (t), was established to determine mean trends and predictions against a background of considerable seasonal and stochastic variability; for the single variables Q explained 0–20 % and T explained 2–58 % of the variation in the concentrations. The relationship was highest for variables in PC2 and lowest for those in PC3 and PC4, where Q and T had little or no influence.

Reasons for rising and falling long-term trends with time are discussed in detail. Austria now contributes only minor proportions to the nutrient load of the Danube, which is causing eutrophication of the Black Sea downstream, and water quality of the Austrian section of the river is good.

The advantages and problems of statistical process analysis are discussed in relation to environmental monitoring programs and the different specific requirements of compliance monitoring.

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Introduction

When a major anthropogenic event occurs, there is often a short-term "impact-study" to discover if the factor has had any harmful effects on a particular system. Unfortunately, a short-term "impact-study" will rarely be of value because it usually tests the null hypothesis of "no change" against an implicit alternative of a "detectable change", usually assumed to be for the worse. Such an approach is often useless because of the problem of assessing the significance of any change when little is known about the spatial and temporal variations in the "base-line" from which the change occurred. Therefore, "impact effects" must be tested against a null-hypothesis error term representing between-years variation in the absence of the "impact".

One major objective of long-term investigations therefore must be to provide reliable estimates of the base-line variation. To ensure that the degrees of freedom for the between-years error term are adequate for statistical tests, regular samples must be taken over many years. Another objective should be to detect any long-term trend in the mean level of the base line. Long-term investigations are also essential for the detection of rare but important events, and for the formulation of meaningful, testable hypotheses. In a multivariate problem, a long-term study is often required to provide the necessary degrees of freedom for statistical analyses (Strayer et al., 1986; Elliott, 1990).

A 39-year series (1957–1995) of data on fourteen physical, chemical and biological variables is now available for the Austrian part of the River Danube west of Vienna-Nußdorf (river-kilometre 1934.7) and the purpose of the present statistical analysis is to detect the long-term trends of these variables in relation to human activities (represented by time), discharge and water temperature.

The Danube, with a length of *c.* 2850 km, is the second largest river in Europe. It rises in the Black Forest (Germany), discharges into the Black Sea (Romania, Ukraine) and crosses six other countries: Slovak Republic, Hungary, Croatia, FR Yugoslavia, Bulgaria and Moldova. The area of the river basin consists of 796 250 km^2 (Gleick, 1993). These figures show the extraordinary international importance of this river.

Changes in the concentration of the measured variables of the River Danube near Vienna, stream-order 9 (Wimmer and Moog, 1994), are inevitably dependent on events occurring in the catchment above a particular water gauge. For the catchment area of the Danube at Vienna-Nußdorf, approximately 55 % lies in the south of Germany (Baden-Württemberg and Bavaria), and only 26 % of the human population in the catchment actually lives in Austria (Table 1).

In the Austrian part of the catchment, in 1968, about 25 % of the population's sewage was discharged into the main river via canalised watercourses and only about 3 % passed through waste-water treatment plants. In 1988, in connection with the completion of the series of hydro-electric power plants on the Danube, sewage treatment was improved along the river, and about 70 % of the sewage was treated before discharge. Similarly, in Germany the treatment of sewage increased from 30 % to 85 % of the population (Table 1). Furthermore, legislation has stepwise decreased polyphosphates from washing powders (Germany in 1980, Austria in 1984) and has improved the technology of cellulose production. Now, legislation has

Table 1. Summary of information on the numbers of people living in different political regions of the catchment areas of the River Danube in Germany, Italy, Switzerland and Austria, in 1968 and 1988 (Humpesch, 1992; Fleckseder, 1994). Also given are the numbers (approximate) of inhabitants connected to canalised sewage systems (CS) and waste-water treatment plants (WTP)

Political region	Catchment-area (km^2)	1968 (millions)			1988* (millions)		
		Population	CS	WTP	Population	CS	WTP
Baden-Württemberg	7580	1.5	0.9	0.3	1.6	1.5	1.4
Bavaria	48690	7.2	4.1	2.7	7.7	6.6	6.4
Italy/Switzerland	2180	<0.1	–	–	<0.1	–	–
Austria	43250	3.1	0.8	0.1	3.2	2.1	1.9
Total at water gauge “Vienna-Nußdorf”	101700	11.8	5.8	3.1	12.5	10.2	9.7

* More recent estimates are not available, because values are given for the areas of the counties and not for the catchment area of the Danube (Fleckseder, pers. comm.).

been passed in both Germany and Austria to oxidise ammonia and reduce the nitrogen content of treated water.

The effects of these measures will be evaluated in the present study, where a time series analysis has been applied for the first time, especially to detect correlations between fourteen physical, chemical and biological variables and their trends related to time, and simulations of trends related to time, discharge and temperature. An attempt will be made to discuss the sources of inputs into the Danube-system, what caused the trends and what will the future trends be. Furthermore the results should give answers for optimising the monitoring of the river system and indicate if and when given water quality values will be met.

Material and methods

Sampling site and data

About 96 % of the Austrian area is drained by the River Danube (BMLF, 1996) and the river shows a complex flow regime with high discharge in early summer and a second short discharge peak in winter (Mader et al., 1996). From Germany upstream, the river enters Austria with a mean discharge of 1430 m^3/s at Achleiten (river-km 2223) and leaves Austria with a mean discharge of 2210 m^3/s at Wolfsthal (river-km 1873) (IWG, 1996). Downstream to Vienna, the Danube is a hyporhithral/epipotamal zone (sensu Illies, 1961; Humpesch, 1989), stream-order 9 (Wimmer and Moog, 1994). From the source of the Danube down to Vienna, 52 power plants in Germany and Austria interrupt the continuum of the river.

In 1995 the biological water quality of the Austrian Danube, measured at the borders with Germany upstream and Slovakia downstream, was in both cases β-mesosaprobic (class II sensu “Münchener Methode” – Liebmann, 1959), although downstream of both Linz and Vienna the water quality index value was β- to α-mesosaprobic (class II–III), with higher bacterial numbers caused by waste-water inputs (BMLF, 1996). In 1996 about 73 % of the Austrian population was connected to biological waste-water treatment, 40% of which incorporated the removal of phosphorus. But concentrations of the measured variables at the Vienna-Nußdorf water gauge are highly influenced by inputs from Germany, because it occupies a large proportion of the upper catchment area and has a large population (Fleckseder, 1994).

The sampling site Vienna-Nußdorf is located at river-kilometre 1934.7 on the right bank where the Danube is entering Vienna (48°16′ N, 16°22′ E, altitude 164 m a.sl.), not influenced by the waste water of the city. Upstream at river-kilometre 1937.6 there is a waste water treatment plant of Klosterneuburg (40 000 population equivalents). Since 1957, a range of physical, chemical and biological variables have been taken at Vienna-Nußdorf by the Institute of Water Quality (Federal Ministry of Agriculture and Forestry). Datasets analysed in this paper span a period of 39 years, from 1957 to 1995. For the period 1957 to 1977, samples were taken once or twice a year, in spring and autumn. In 1978 (1981 for bacteria) to 1995, sampling was usually monthly at comparable daytime. Details of the number of samples, arithmetic mean values and standard deviations of the data, are given in Appendix I. Table 2 lists the 16 variables and abbreviations used for statistical analysis.

Explorative data analysis

Descriptive parameters were calculated for each year and month: arithmetic means, standard deviations for annual values (1957 onwards, Appendix I), plus coefficients of variation and 95%-confidence intervals for monthly values (1978 onwards, see Appendix II). Shapiro-Wilks'-W test was used for testing the hypothesis of normal distribution; non-normally distributed data were $\log_{10}$-transformed. Outliers were determined by applying Grubbs-test (Hartung et al., 1993). In all statistical tests the level of significance α was 0.05.

Table 2. Sixteen variables from the River Danube, used for statistical analysis. “Data available” indicates the first year in which measurements began. CFU = colony-forming units

Variable Y (units)	Abbreviation	Data available	Observations
Discharge (m^3/s)	Q	1957	252
Temperature (°C)	T	1957	253
Chloride (mg/l)	Cl	1957	243
Nitrate (mg N/l)	NO_3	1957	245
Nitrite (mg N/l)	NO_2	1957	238
Ammonium (mg N/l)	NH_4	1957	240
Soluble reactive phosphorus (mg P/l)	SRP	1957	245
Total phosphorus (mg P/l)	TP	1978	219
Potassium permanganate (mg/l)	$KMnO_4$	1957	246
5-day biochemical oxygen demand (mg O_2/l)	BOD5	1971	231
2-day biochemical oxygen demand (mg O_2/l)	BOD2	1957	243
2-day biochemical oxygen demand (%)	BOD2%	1957	243
Dissolved oxygen (mg O_2/l)	O_2	1957	247
Dissolved oxygen (% saturation)	O_2%	1957	241
Heterotrophic saprophytic bacteria (CFU/ml)	HB	1957	203
Faecal coliforms (CFU/ml)	FC	1957	202

Explorative multivariate analysis was done by principal component analysis (PCA) with normalised varimax-rotation (Backhaus et al., 1993). The criterion for extracting a principal component was an Eigen-value greater than 1.

Trends related to time

Independent variable time t was treated as: t = (metric year – 1900). For initial identification of the chronological sequences of observations, the following models were used.
Polynomial model for smooth trends (S) in time:

$$Y_S = b_{0\,1} + [b_1\,t] + [b_2\,t^2] \tag{1}$$

Sinusoid model incorporating a cyclic component (C):

$$Y_C = b_{0\,2} + [b_3 \sin(2\pi f)] + [b_4 \cos(2\pi f)] \tag{2}$$

Combined model for trends in time (Tr):

$$Y_{Tr} = Y_S + Y_C \tag{3}$$

where f is the sampling date calculated as: (days of the year)/365, $b_{0\,1} \ldots b_5$ are estimated coefficients (Appendix III and IV), and Y is a dependent variable.

The coefficients for all models were estimated by multiple linear regressions, least squares method. The measure of goodness-of-fit was the coefficient of determination R^2. Initially the time series were fitted to model (1). To make the time series stationary for applying model (2), significant smooth components were removed from data and residuals from 1978 onward were processed in Fourier (spectral) analysis to detect cyclic fluctuations. Periodogram values were smoothed by symmetric Tukey-Hanning-window for 5 points (Schlittgen and Streitberg, 1994). The final time-trend model (3) is a combination of models (1) and (2).

Simulation of trends related to time, discharge and temperature

For attaining fast information from easily measurable variables, variables (Y) were individually regressed against two independent environmental variables, discharge (Q) and temperature (T), using quadratic equation (1). The variables were also related to these easy measurable variables combined in a multiple polynomial equation:

$$Y = b_{0\,4} + [b_1\,t] + [b_2\,t^2] + [b_3\,T] + [b_4\,T^2] + [b_5\,Q] + [b_6\,Q^2] \tag{4}$$

where $b_{0\,4} \ldots b_6$ are estimated coefficients; the coefficients of determination (R^2) were estimated by multiple regressions (Appendix V).

"Optimised" models were calculated for equations (1) to (4); significant coefficients were found by stepwise regression ($p_{in} = 0.05$, $p_{out} = 0.10$).

Results

Data structure

All variables (Table 2) except discharge, temperature and soluble reactive phosphorus (SRP) were $\log_{10}$-transformed (lg). A preliminary PCA including two independent variables (discharge and temperature) and 14 dependent variables, was employed to reduce the number of the latter in further analyses. Six principal components (PC) explaining 59 % of total variance were extracted, in which temperature was negatively associated with chloride, nitrate and oxygen. Variable discharge as well as oxygen (%) did not fit any of the extracted components. PC2 showed that 5-day and 2-day biochemical oxygen demand (BOD5, BOD2 and BOD2 (%)) are highly correlated. Consequently, three variables – oxygen (%), BOD2, and BOD2 (%) – were removed from further analysis to reduce variability and redundancy.

A second PCA with the remaining 11 dependent variables showed four main components explaining 72 % of total variance (Table 3, Fig. 1). Group PC1 contains total phosphorus (TP), $KMnO_4$ (oxidation of dissolved organic carbon by potassium permanganate) and SRP, PC2 contains nitrate, chloride and oxygen, PC3 contains BOD5 and nitrite, and PC4 contains bacteria. Ammonium and nitrite are not explained satisfactorily (for each group < 50 %) but have their main proportions in PC1 and PC3, respectively.

Trend analyses

Except for ammonium, variables in PC1 show a high affinity to trend in time during the 39-year period 1957 to 1995, explaining 36 to 58 % of individual variation (Table 4). The time-trends of these variables may be represented by quadratic curves, rising from low values in the 1950s and 1960s to peak during the 1970s and 1980s, followed by steady declines through the early 1990s (for mean values see Appendix I). The most pronounced curve was for SRP (Fig. 2). Similarly, variables

Table 3. Results from PCA after normalised varimax-rotation, showing four groupings of 11 selected measured variables from the River Danube (for abbreviations see Table 2). PC1 to PC4 are principal components 1 to 4 (see Fig. 1). Bold numbers indicate loadings > 0.6

Variable	PC1	PC2	PC3	PC4	Communality
Total phosphorus	**0.935**	0.091	0.143	0.057	90.7
$KMnO_4$	**0.801**	−0.064	−0.304	0.271	81.2
SRP	**0.759**	0.260	0.409	0.021	81.1
Ammonium	**0.625**	0.460	0.109	−0.100	61.8
Nitrate	0.161	**0.862**	0.189	0.062	80.8
Chloride	0.309	**0.829**	0.167	−0.088	81.9
Dissolved oxygen	−0.072	**0.787**	−0.357	−0.073	75.2
BOD5	0.010	0.083	**−0.821**	0.027	68.1
Nitrite	0.129	0.126	**0.641**	0.093	45.2
Heterotrophic bacteria	0.137	0.182	0.073	**0.788**	67.8
Faecal coliforms	0.035	−0.249	0.006	**0.754**	63.1
Variance explained %	24.1	22.4	14.3	11.8	72.5

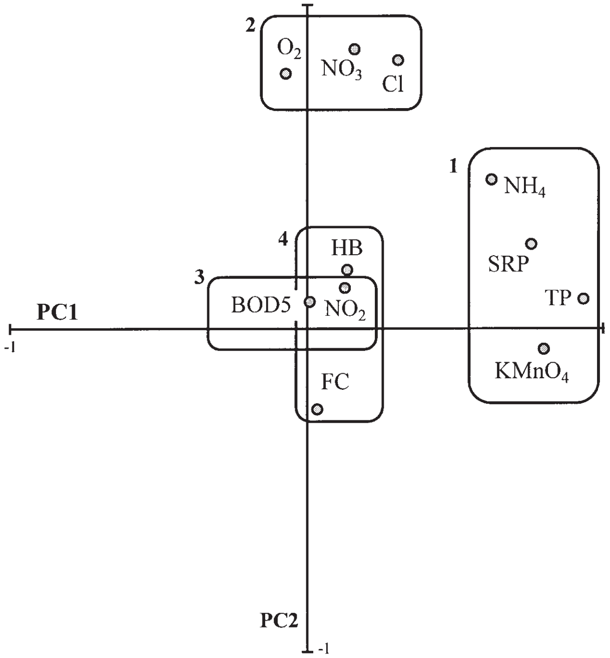
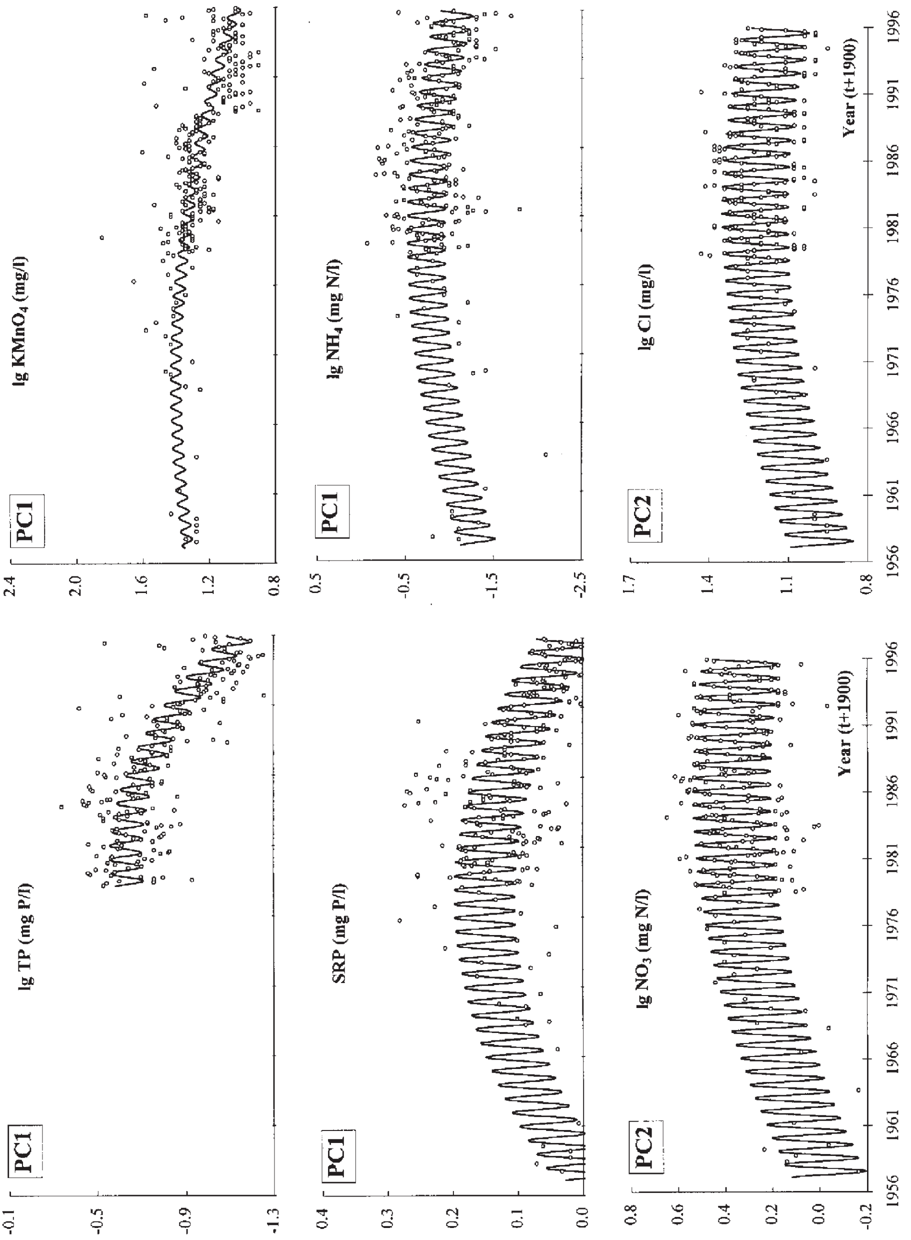


Figure 1. PCA on 11 selected variables from the River Danube. Axes PC1 and PC2 represent principal components 1 and 2, bold numbers (1 to 4) indicate variable-clusters (for abbreviations see Table 2)

Table 4. Model equations for trends in time, applied to physical, chemical and biological variables of the River Danube (for abbreviations see Table 2): polynomial smooth-trend model (1), sinusoid cycle (2) plus minimal and maximal values (Min, Max; Appendix II) and a combined total time-related model (3). R^2 is the adjusted coefficient of determination, with levels of significance (Sign.) *** $p < 0.001$, ** $p < 0.01$, * $p < 0.05$, o $p \geq 0.05$. "Optimised" values were obtained from model (3) using significant coefficients only (see Methods)

PC	Variable	Model							
		(1) Trend S		(2) Cyclic C				(3) "Optimised" Tr	
		R^2	Sign.	R^2	Sign.	Min	Max	R^2	Sign.
–	Discharge	0.000	o	0.313	***	Nov	Jun	0.313	***
–	Temperature	0.000	o	0.909	***	Jan	Aug	0.912	***
1	Total phosphorus	0.579	***	0.140	***	Jul	Mar	0.638	***
1	$KMnO_4$	0.421	***	0.036	**	Nov	Oct	0.442	***
1	SRP	0.365	***	0.418	***	May	Jan	0.632	***
1	Ammonium	0.149	***	0.247	***	May	Feb	0.361	***
2	Nitrate	0.222	***	0.587	***	Jul	Feb	0.686	***
2	Chloride	0.127	***	0.681	***	Jul	Feb	0.726	***
2	Dissolved oxygen	0.086	***	0.573	***	Aug	Feb	0.618	***
3	BOD5	0.001	o	0.075	***	Nov	Apr	0.075	***
3	Nitrite	0.014	o	0.033	**	Apr	Nov	0.033	***
4	Heterotrophic bacteria	0.000	o	0.049	**	Aug	Jan	0.045	**
4	Faecal coliforms	0.000	o	0.096	***	May	Oct	0.096	***


PC1
lg KMnO$_4$ (mg/l)
2.4
2.0
1.6
1.2
0.8
PC1
lg TP (mg P/l)
-0.1
-0.5
-0.9
-1.3
PC1
lg NH$_4$ (mg N/l)
0.5
-0.5
-1.5
-2.5
PC1
SRP (mg P/l)
0.4
0.3
0.2
0.1
0.0
PC2
lg Cl (mg/l)
1.7
1.4
1.1
0.8
PC2
lg NO$_3$ (mg N/l)
0.8
0.6
0.4
0.2
0.0
-0.2
1956
1961
1966
1971
1976
1981
1986
1991
1996
Year (t+1900)

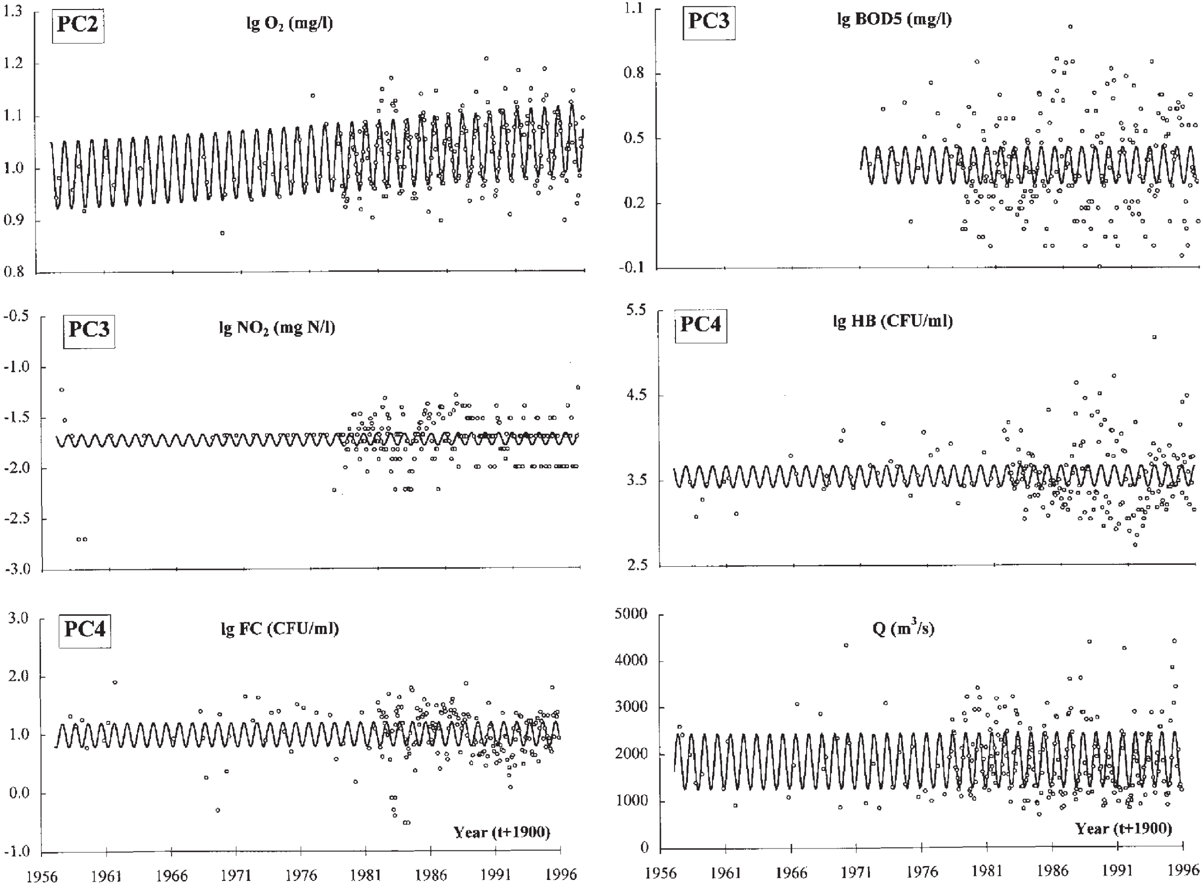


Figure 2. Models for 12 variables in the River Danube, for sampling periods spanning up to 39 years (1957 to 1995). The seasonal (annual) curves are calculated from "optimised" time-trend model (3) (for abbreviations see Table 2)

in PC2 showed weaker but significant trends over the same time-period, whereas those in PC3 and PC4 did not (Fig. 2 and Table 4).

All analysed variables have significant cyclic fluctuations, and for most of them spectralplots revealed a periodicity of 12 months, indicating seasonal variation. In fact, sinusoid model (2) showed that an annual cyclic component explained 57 to 68% of the variability for group PC2, and 4 to 42% for group PC1 (Table 4); minimum concentrations occurred in summer and maximum values occurred in winter. Variables in PC3 and PC4 also have significant though weak relationships to annual cycles. The annual cyclic components around long-term trends for the 11 selected variables and discharge are illustrated in Figure 2.

When fourteen measured variables were individually regressed against the two independent variables, discharge (Q) and temperature (T), values for the coefficients of determination (R^2) obtained from polynomial model equation (1) showed that Q explained only 0 to 20%, whereas T explained 2 to 58% of the variation in concentrations (Table 5, Appendix IV). The relationships for both variables over the 39-year study period were highest for group PC2, and they were lowest for groups PC3 and PC4 with minor or no influence of temperature and discharge.

For simulations using the combined model (4), which includes the independent variables discharge, temperature and time, the values for R^2 ranged from 42 to 72% for groups PC1 and PC2, and 60% or more of the variances were explained for five variables (TP, $KMnO_4$, SRP, NO_3 and Cl) using model (4) (Table 5). By comparison with values of R^2 obtained from model (1) in Table 4 it is evident that the main proportion of the variance (i.e., variation in concentrations) for group PC1 was related to time, whereas for variables grouped in PC2, variation was mainly related to temperature (model (1) in Table 5).

Table 5. Values of the adjusted coefficient of determination (R^2) with significance (Sign.: *** $p < 0.001$, ** $p < 0.01$, * $p < 0.05$, o $p \geq 0.05$), for 11 selected variables from the River Danube in four PC groupings (for abbreviations see Table 2). R^2 was obtained from polynomial model (1), with discharge and temperature as separate independent variables, and from multiple polynomial model (4) with three variables combined (time, discharge and temperature). "Optimised" values were obtained from model (4) using significant coefficients only (see Methods)

PC	Variable	Model (1)				Model (4)			
		Discharge Q		Temperature T		Combined		"Optimised"	
		R^2	Sign.	R^2	Sign.	R^2	Sign.	R^2	Sign.
1	Total phosphorus	0.003	o	0.091	***	0.693	***	0.693	***
1	$KMnO_4$	0.072	***	0.069	***	0.598	***	0.600	***
1	SRP	0.132	***	0.202	***	0.630	***	0.629	***
1	Ammonium	0.021	*	0.271	***	0.421	***	0.419	***
2	Nitrate	0.144	***	0.507	***	0.689	***	0.681	***
2	Chloride	0.204	***	0.576	***	0.720	***	0.720	***
2	Dissolved oxygen	0.031	**	0.383	***	0.470	***	0.477	***
3	BOD5	0.000	o	0.018	o	0.007	o	0.020	*
3	Nitrite	0.016	o	0.043	**	0.065	**	0.041	**
4	Heterotrophic bacteria	0.050	**	0.034	*	0.087	***	0.094	***
4	Faecal coliforms	0.000	o	0.030	*	0.038	*	0.035	**

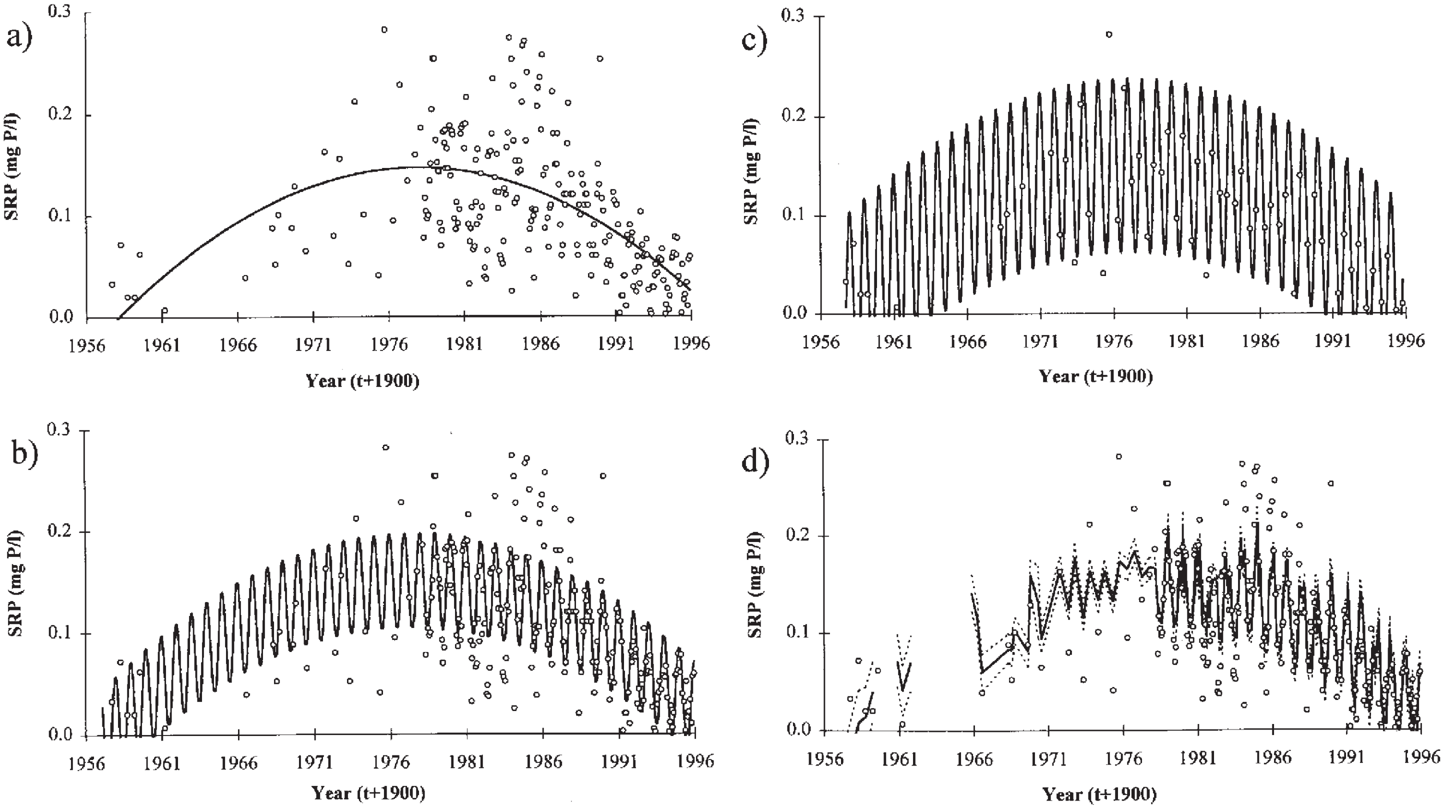


Figure 3. Concentrations of SRP in the River Danube during the period 1957 to 1995: a) Smooth-trend model (1), b) Time-trend model (3) for all values, c) Time-trend model (3) for sampling twice a year (spring and autumn), d) "Optimised" multiple polynomial model (4) incorporating time, discharge and temperature with 95%-confidence intervals (broken lines)

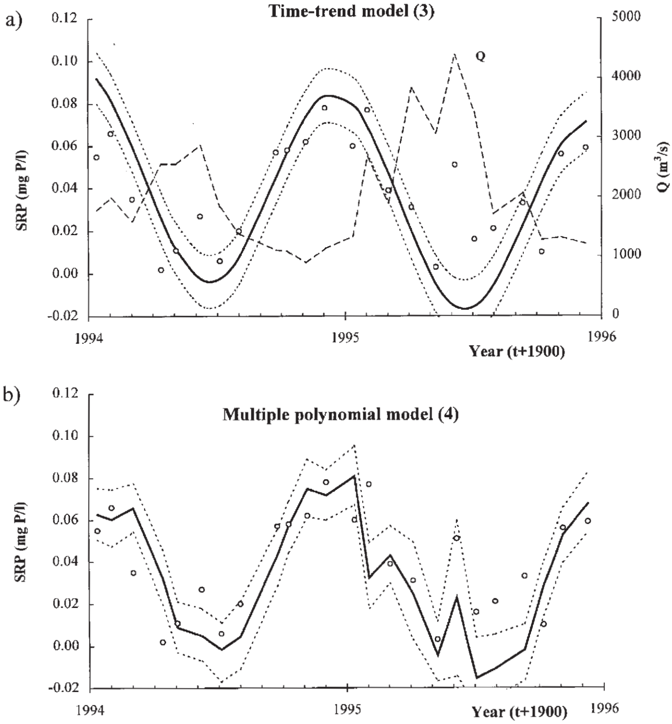


Figure 4. Concentrations of SRP obtained monthly from the River Danube in 1994 and 1995, with "optimised" curves and 95%-confidence intervals: a) Time-trend model (3) and discharge (Q, broken line), b) Multiple polynomial model (4)

The concentrations of SRP are taken as an example for the conclusion of the statistical analysis (Fig. 3) and plotted against the "optimised" models (1), (3), and (4). Model (3) used for monthly values explains 63% of the variance (Fig. 3b), but two values a year (spring and autumn) give an even better explanation of 70% of the variance (Fig. 3c). Model (4) generated annual cyclic fluctuations in SRP, closely resembling those generated by the time-trend model (3). The interesting point is that the calculation according to model (4) gives a very good prediction after 1986, and the reason for this will be discussed later.

The usefulness of the models is illustrated in Figure 4, which presents a 2-year plot of SRP concentrations obtained each month in 1994 and 1995, together with "optimised" time model (3) and "optimised" model (4). Model (3), with independent variable time only, gives a reasonable description of annual fluctuations in SRP. However, model (4), which incorporates discharge and temperature as well as time, gives a better description of single outlying and stochastic events for concentrations of SRP, which implies a good predictive capability.

Discussion

River Danube in Austria

In the present study seven out of eleven variables at Vienna-Nußdorf were extracted by PCA into two main components or groups with different causes.

The first group (PC1) of four variables (total phosphorus, soluble reactive phosphorus, ammonium and potassium-permanganate) predominantly represents "point-source inputs" that are strongly influenced by corrective measures in the catchment, and time series analysis shows the results of successful waste-water management. Thus, the second order (quadratic) polynomial model (1) reveals pronounced increasing long-term trends in concentrations of all four variables from the 1950s up to the 1970s. These were followed by decreasing trends in the 1980s and 1990s (Fig. 2), corresponding to a reduction of phosphate-use in detergents and improved waste-water treatment in the catchment (Petto et al., 1991 a). For example, for the whole of Austria, estimated inputs of phosphorus derived from the use of detergents fell from 5300 t P in 1981 to only 525 t P in 1992. In the same period, total inputs of phosphorus derived from waste-water after treatment in Austria, fell from 8740 t P to 3700–4300 t P, whereas the nonpoint phosphorus input from agricultural areas remained constant at 8400 t P (Fleckseder, 1987; IWA, 1997), indicating a shift from point-sources to nonpoint-sources.

The overall effect of these management measures since the 1980s has been to reduce the average concentrations of TP and SRP by more than half in the Austrian section of the Danube, and NH_4 and $KMnO_4$ are currently about half the peak concentrations of the 1980s (Appendix I).

Three variables in the second group (PC2) also exhibit increasing long-term trends in concentrations during the 1950s to 1970s, but those for the variables nitrate and chloride levelled off from the beginning of the 1980s onwards (Hadl and Kreitner, 1991); they now begin to show decreasing trends in the 1990s. Nitrogen inputs to the Danube are mainly from agricultural fertilisers and fuel incineration (Fleckseder and Zeßner-Spitzenberg, 1994); about 60 % is from atmospheric deposition and agriculture (Schwaiger, 1994). In 1992 compared to 1988, 5 % lower load-values were assumed for nitrogen caused by reduced fertiliser-application (IWA, 1997). The oxygen content of the river has steadily increased since the 1950s (Fig. 2), indicating an improving water quality in spite of trendless chlorophyll-*a* concentrations (Rodinger, 1991). In contrast to group PC1, variables in group PC2 appear to be more strongly related to river discharge and temperature (Model (1), Table 5), suggesting they are more directly influenced by seasonal (annual) patterns of Q and T.

However, the overall mean monthly concentrations of all variables in both PC1 and PC2, except oxygen, reached maxima in winter and minima in summer, when concentrations were approximately halved (Appendix II). On the other hand, river discharge reached maximal values in summer and minimal in winter, when values were approximately half those of mid summer. Therefore dilution in summer and concentration in winter must account for a large part of the pronounced annual cycle of fluctuations in the variables of PC1 and PC2; Figure 4 illustrates an example of the annual inverse relationship between SRP concentrations and Q. The exception is oxygen, because its solubility in water is strongly temperature-dependent. Thus annual fluctuations in oxygen concentration (mg/l) related to T would be expected, being high in winter and low in summer. In contrast, mean monthly values for % oxygen varied little throughout the year (Appendix II), indicating that the river is fully saturated with oxygen at all temperatures, even during mid summer when, in fact, the river is slightly super-saturated. Variability in concentrations of the other variables in PC1 and PC2 is partly dependent on T through the latter's dominating effect on metabolic processes, both within the river and on the catchment.

Variables in group PC2, even chloride in winter, may be considered as mainly "nonpoint-sources or diffuse inputs", ruled or driven by seasonal factors induced by different mechanisms; for nitrate these are mainly leaching losses from soil, as discussed in MacDonald et al. (1994). Long-term trends for further reductions in chloride and nitrate will depend on management decisions made in Austria, but also upstream in Germany. Nitrate concentrations in particular may continue to fall as de-nitrification processes are introduced at waste-water treatment plants.

The third and fourth PCA components are characterised as groups of variables that do not exhibit smooth trends in time, i.e. concentrations of the variables did not change over the 39 years of study, although there are weak annual cycles around the mean values. Bacteria form a single component (PC4) as in other studies of the Austrian part of the River Danube (Petto et al., 1991 b; Humpesch, 1991). Surprisingly, anthropogenic BOD5, as well as the numbers of faecal bacterial, seem to remain constant throughout the study period, despite the introduction of several biological waste-water treatment plants along the Austrian section of the Danube, e.g., the starting of the biological waste-water treatment at Klosterneuburg in 1989 at river-km 1937.6, only a few kilometres above the sampling site at Vienna-Nußdorf. BOD5 does not show any quality changes in the River Danube. Descriptive values for faecal bacteria indicate a shift to lower numbers in the period 1989 to 1994, similar to $KMnO_4$, but for analysis this fact is overwhelmed by the high values at high discharges in 1995 (Appendix I).

From the results presented in this study, it is clear that the following measures had a significantly positive effect on the water quality of the Danube in the last ten years: improvements in waste-water treatment, increasing from 3% biological treatment in 1968 to 73.5% in 1995 (Emde, 1970; BMLF, 1996); substitution of phosphate in detergents begun by the industry in 1974 (Müller, 1996), was accompanied by stepwise governmental threshold-values in Switzerland (1977), Germany (1981 and 1984), and Austria (1985 and 1987); reduction in emissions of pulp and paper mills; intensive and widespread monitoring (e.g., Austrian water quality monitoring system, Chovanec et al., 1996) accompanied by governmental laws such as the revision to the Austrian Water Act 1959 (Republik Österreich, 1959 and 1990) and threshold values. Furthermore, by introducing the term "ecological capability" into the Austrian Water Act a sensible approach was made to maintain the natural functioning of an aquatic habitat (Chovanec et al., 1994).

Statistical process analysis and general implications for monitoring water quality

In the past, objectives of an assessment were not always exactly defined (Chapman, 1996) and variables were sometimes taken arbitrarily (Meybeck et al., 1989). Thus the currently available multivariate datasets usually contain much information that is useful but also some that is redundant. The application of statistical methods is necessary in order to abstract and present the information concisely or, as stated in Green and Montagna for benthic communities (1996), the "design of a monitoring study and statistical approaches to interpretation of results are inseparable", which is true for all monitoring studies.

The ordination of datasets by PCA helps to uncover underlying factors (components) and groups of related variables. Each group contains variables with similar patterns, related to a "ruling" or "driving" factor, inferring that the variables have or may have the same background influence or interdependencies. Therefore a particular variable in a group that is of interest can be chosen as a so-called "key-variable" to represent the whole group, and thus reduce the dataset of the monitoring program to a minimum of requirements. For example, group PC1 for the Danube contains variables that are considered to be strongly influenced by a combination of "point-source inputs" and "successful management", and currently this group can be represented by concentrations of the key-variable "soluble reactive phosphorus (SRP)". Of course one has to keep in mind that, as in this study variables with different causes and mechanisms may be combined in a single group so one has to be very careful when excluding variables for further monitoring. However, as shown here, it is possible to omit some, e.g. BOD2, without loss of information. In general, environmental programs with enhanced datasets need additional analysis by ordination to detect underlying processes or relationships. Carefully considered reductions of a given set of variables can improve the sampling strategy for monitoring in a cost-effective manner. The money saved may be used to increase the number of measurements made on interesting "key-variables", and this leads to both higher precision and higher accuracy of estimates, descriptions and predictions obtained from mathematical and statistical models.

Monitoring has to provide basic information about temporal and spatial variations of values (base-line limits) and should be able to detect changes within set limits and, if possible, identify their causes (Parr, 1996). The first thing to do when searching for pattern in time is to view descriptive plots of the variables of interest in order to get a first idea of temporal variation. Time series analysis now constitutes a powerful tool for identifying phenomenona such as trends and cycles, and it reveals "the invisible present" (Magnuson, 1995). The detection of underlying patterns requires sampling periods that are regular or equally spaced in time, e.g., biweekly or monthly as in this study since 1978. As shown, it is possible to relate annual fluctuations of single variables, or even groups of variables, to internal and external processes on the catchment or in the river. Identified base-line values for each variable make it possible to reveal sudden or progressive changes in the values, or violation of established concentration limits, or to predict expectation of seasonal maxima and minima.

At the start of a routine monitoring program, sampling should be very intensive for both time intervals (frequency) and even sampling size (numbers of samples and (preferably) several subsamples from the river). Once the base-line for each variable has been properly established, normally it should then be possible to reduce the sampling frequency (but not necessarily the number of samples at each visit). An example is given in Figure 3c, where only the values for SRP in March and September were employed for comparison with the analysis of all monthly values, shown in Figure 3b. This was done to exclude high or low concentrations of SRP associated with irregular disturbances on the catchment and in the river, such as snowmelt in early summer and stochastic spates mainly in late autumn or winter. Although the greatly reduced dataset produced (as expected) more pronounced cyclic variations around the mean (Fig. 3c), regression analysis with model (3) explained 70% of the total variance, compared with 63% for all monthly values (Fig. 3b).

Problems may arise when establishing a regression model over a long time period: On one hand it should cover the whole data-set and on the other hand it should remain as simple as possible. The conflict is shown in Fig. 4, where real values keep on the same low level but modelled values become negative, which is a tribute to the second order polynomial model.

Environmental fluctuations naturally cause periodic, seasonal and annual variations in measured variables (e.g., Webb and Walling, 1992), but some may result from other changes within and between years. If monitoring programs would generally consider more fundamental patterns in variation, the gain could be better information about the system being studied (Hakanson and Peters, 1995). Only trend and spectral analysis of long-term data can give answers to the following questions: do the measurements represent a long-term development, and is there a local input or were measurements done at unfavourable times? A further important point for specific monitoring programs is the number of measurements required for analysing data against a certain threshold: did the monitoring program actually provide the opportunity to detect violation of certain limits (Type II error)? A sufficient number of measurements can be attained only by prior determination of the accuracy and power of the statistical test that is to be applied for evaluating specified differences between monitored values.

Reliable statistical models provide forecasts of expected concentrations or other values, and the observed values can be compared with predictions. This procedure can be used to assess the sufficiency and efficiency of management programs or indicate requirements for immediate corrective measures. Statistical analyses are not a sophisticated luxury but necessary tools for the identification of base-lines – in this study, means, seasonality and long-term trends – which can be applied to optimise the sampling strategy required for routine monitoring, to identify important "ruling" or "driving" components, and for further prognosis. Consequently the analyses provide policymakers with both strategic and tactical decision support.

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Appendix I. Statistics for yearly values of 16 variables sampled in the River Danube during the period 1957 to 1995. For abbreviations see Table 2; Σ = sum, avg = arithmetic mean, std = standard deviation, n = number of observations.

	Q (m^3/s)			**T** (°C)			**Cl** (mg/l)			**NO_3** (mg N/l)			**NO_2** (mg N/l)			**NH_4** (mg N/l)			**SRP** (mg P/l)			**TP** (mg P/l)		
year	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n
57	2504	121	2	13.1	1.4	2	-	-	0	0.460	0.325	2	0.045	0.021	2	0.117	0.054	2	0.033	-	1	-	-	0
58	1701	429	2	10.9	4.4	2	9	0	2	1.323	0.081	2	0.012	0.013	2	0.063	0.022	2	0.046	0.037	2	-	-	0
59	1583	-	1	12.0	7.6	2	10	0	2	1.323	0.569	2	0.002	-	1	0.093	0.000	2	0.041	0.030	2	-	-	0
60	1280	-	1	7.4	-	1	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
61	1602	964	2	11.9	5.7	2	12	-	1	1.288	-	1	0.021	-	1	0.039	-	1	0.007	-	1	-	-	0
63	-	-	0	14.6	-	1	9	-	1	0.690	-	1	0.021	-	1	0.008	-	1	-	-	0	-	-	0
65	1090	-	1	5.5	-	1	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
66	2423	930	2	10.9	7.1	2	-	-	0	1.200	-	1	-	-	0	-	-	0	0.039	-	1	-	-	0
68	2168	619	3	15.5	4.1	3	12	2	3	1.303	0.479	3	0.020	-	1	0.101	-	1	0.080	0.025	3	-	-	0
69	1606	1042	2	14.8	4.3	2	17	0	2	1.380	0.325	2	0.021	0.000	2	0.047	0.011	2	0.109	0.029	2	-	-	0
70	3288	1489	2	13.1	4.4	2	10	-	1	2.070	-	1	0.021	-	1	-	-	0	0.065	-	1	-	-	0
71	958	-	1	11.8	-	1	16	-	1	2.760	-	1	0.021	-	1	0.078	-	1	0.163	-	1	-	-	0
72	1326	661	2	12.0	1.8	2	17	2	2	2.070	0.325	2	0.021	-	1	-	-	0	0.118	0.054	2	-	-	0
73	2190	1268	2	10.3	1.6	2	18	1	2	2.415	0.163	2	0.021	-	1	0.233	0.219	2	0.132	0.113	2	-	-	0
74	1698	480	2	9.8	1.3	2	15	4	2	2.070	0.651	2	0.021	0.000	2	0.128	0.093	2	0.101	-	1	-	-	0
75	1616	760	2	10.7	0.1	2	15	2	2	2.530	0.651	2	0.021	-	1	0.116	0.000	2	0.162	0.170	2	-	-	0
76	1104	148	2	10.0	1.6	2	16	2	2	2.530	0.325	2	0.021	0.000	2	0.194	0.055	2	0.162	0.094	2	-	-	0
77	1435	429	2	9.7	2.9	2	18	0	2	2.703	0.732	2	0.014	0.011	2	0.194	0.055	2	0.147	0.018	2	-	-	0
78	1905	507	10	11.2	5.2	10	17	4	10	2.031	0.600	10	0.018	0.004	10	0.156	0.067	10	0.143	0.056	10	0.228	0.072	10
79	2023	728	13	10.1	4.9	14	16	5	14	2.294	0.659	14	0.023	0.007	13	0.204	0.095	13	0.151	0.047	14	0.254	0.050	13
80	2167	791	12	9.0	5.8	12	17	3	12	2.367	0.678	12	0.022	0.007	12	0.252	0.137	12	0.145	0.043	12	0.225	0.060	12
81	2040	591	14	10.6	6.1	14	17	4	14	2.190	0.817	14	0.024	0.010	14	0.170	0.160	14	0.116	0.054	14	0.220	0.052	14
82	1936	664	12	10.5	6.5	12	16	4	12	2.206	0.676	12	0.018	0.010	12	0.174	0.106	12	0.119	0.064	12	0.213	0.059	12
83	1983	773	13	10.0	5.6	13	17	4	13	1.920	0.631	13	0.012	0.006	13	0.193	0.108	13	0.114	0.043	13	0.267	0.087	13
84	1568	497	13	9.9	5.5	13	17	5	13	2.370	0.864	13	0.027	0.007	13	0.239	0.181	12	0.173	0.071	13	0.260	0.068	13
85	1791	728	12	10.1	6.2	13	17	5	12	2.486	0.794	12	0.024	0.012	12	0.291	0.198	12	0.140	0.074	13	0.226	0.067	12
86	1584	682	12	10.7	6.6	12	19	4	12	2.867	0.866	12	0.025	0.012	12	0.283	0.176	10	0.153	0.062	12	0.220	0.050	12
87	2105	859	12	9.6	5.9	12	17	5	12	2.833	0.691	12	0.027	0.010	12	0.176	0.114	12	0.130	0.045	12	0.205	0.050	12
88	2217	1054	12	10.6	5.9	11	17	4	11	2.534	0.693	11	0.022	0.008	11	0.177	0.109	11	0.102	0.037	11	0.151	0.044	10
89	1862	572	12	10.4	5.7	12	15	3	12	2.327	0.668	12	0.021	0.008	12	0.169	0.073	12	0.095	0.035	12	0.146	0.025	12
90	1429	356	11	10.9	6.5	12	16	3	12	2.428	0.792	11	0.020	0.007	11	0.189	0.129	11	0.095	0.061	11	0.187	0.084	11
91	1654	997	12	10.2	6.8	12	19	5	11	2.505	0.866	11	0.018	0.010	11	0.168	0.089	12	0.057	0.039	12	0.116	0.049	12
92	1626	599	12	11.9	7.1	12	15	4	12	2.174	0.828	12	0.019	0.007	12	0.133	0.077	12	0.057	0.023	12	0.105	0.020	12
93	1772	530	12	10.6	6.9	12	15	4	12	2.158	0.696	12	0.018	0.006	12	0.063	0.018	12	0.040	0.022	12	0.089	0.023	12
94	1733	657	12	11.5	6.3	12	15	4	12	2.392	0.700	12	0.017	0.008	12	0.098	0.038	12	0.040	0.026	12	0.077	0.017	12
95	2350	1103	12	9.9	6.0	12	14	4	12	2.342	0.761	12	0.022	0.015	12	0.116	0.104	12	0.038	0.023	12	0.118	0.064	12
Σ			249			253			243			245			237			236			244			216

Appendix I (continued)

	KMnO$_4$ (mg/l)			**BOD5** (mg O$_2$/l)			**BOD2** (mg O$_2$/l)			**BOD2** (%)			**O$_2$** (mg/l)			**O$_2$** (%)			**HB** (CFU/ml)			**FC** (CFU/ml)		
	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n	avg	std	n
57	21	2	2	-	-	0	0.6	0.0	2	7	0	2	9.2	0.6	2	92	2	2	-	-	0	-	-	0
58	19	0	2	-	-	0	0.6	-	1	7	-	1	9.1		1	93	-	1	2150	1344	2	17.5	4.9	2
59	23	6	2	-	-	0	1.2	-	1	12	-	1	9.2	1.3	2	90	2	2	2400	707	2	12.0	8.5	2
60	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	3100	-	1	8.0	-	1
61	24	-	1	-	-	0	1.5	0.8	2	14	7	2	9.9	0.8	2	96	3	2	2350	1485	2	48.0	45.3	2
63	19	-	1	-	-	0	1.1	-	1	11	-	1	10.0	-	1	103	-	1	-	-	0	-	-	0
65	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	6200	-	1	-	-	0
66	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	3800	-	1	8.4	-	1
68	20	3	2	-	-	0	1.6	0.8	3	17	8	3	9.6	0.8	3	101	10	3	3023	535	3	11.7	11.8	3
69	28	1	2	-	-	0	1.3	0.2	2	15	1	2	8.2	1.0	2	85	2	2	10750	2051	2	11.1	14.9	2
70	20	-	1	-	-	0	1.8	-	1	19	-	1	9.5	-	1	101	-	1	3040	651	2	5.7	4.8	2
71	27	-	1	2.4	-	1	1.2	-	1	14	-	1	8.7	-	1	85	-	1	4750	-	1	44.0	-	1
72	34	6	2	3.5	1.2	2	1.6	0.4	2	16	4	2	10.1	0.1	2	99	4	2	9295	7700	2	29.5	17.7	2
73	30	5	2	2.6	0.3	2	1.3	0.2	2	13	1	2	9.3	0.6	2	88	9	2	4950	495	2	15.4	10.7	2
74	26	1	2	3.0	2.3	2	2.0	2.1	2	20	21	2	9.7	0.4	2	91	6	2	2600	707	2	17.8	9.5	2
75	25	4	2	2.8	0.6	2	1.5	0.2	2	14	0	2	10.3	1.4	2	98	13	2	7600	5657	2	18.5	19.1	2
76	28	-	1	4.9	1.1	2	2.7	0.8	2	23	2	2	11.7	2.9	2	110	32	2	6700	707	2	28.0	-	1
77	25	7	2	2.9	0.4	2	1.8	0.1	2	16	3	2	10.8	1.8	2	100	10	2	5750	3889	2	23.0	-	1
78	22	4	10	2.1	0.8	10	1.0	0.4	10	10	4	10	9.5	1.1	10	91	10	10	2200	707	2	12.4	12.2	2
79	23	3	13	2.6	1.6	13	1.5	1.2	14	11	4	13	10.4	1.0	14	96	12	13	2900	-	1	6.8	-	1
80	22	4	12	2.1	0.8	12	1.0	0.4	11	10	4	11	10.3	1.3	12	93	6	12	5000	2404	2	12.3	15.2	2
81	20	5	14	2.7	1.0	14	1.4	0.8	13	13	6	13	11.1	1.7	14	102	12	13	7350	6576	2	8.8	4.5	2
82	19	2	12	2.2	0.8	12	1.0	0.5	12	9	4	12	11.4	2.0	12	106	13	12	5317	3693	12	18.1	13.2	12
83	20	3	13	2.0	0.6	13	1.5	1.3	13	11	4	12	10.7	1.3	13	99	10	13	4500	2873	13	15.1	13.0	13
84	20	2	13	2.7	1.4	12	1.6	1.2	13	14	10	13	11.2	1.7	13	104	16	11	3008	1006	12	13.6	20.7	12
85	22	6	13	3.3	2.0	13	1.7	1.3	13	14	8	12	10.7	1.6	13	96	10	11	3992	5390	12	21.8	8.0	12
86	20	3	12	3.6	2.2	11	1.7	1.4	10	18	15	11	10.7	1.0	12	99	13	11	2667	1452	12	13.9	7.9	12
87	19	3	12	2.0	1.0	11	1.0	0.4	11	10	4	11	10.8	1.4	12	98	8	12	11083	13233	12	22.2	10.9	12
88	15	7	11	2.6	1.3	11	1.6	0.8	11	14	7	11	11.3	2.3	11	105	13	11	8645	10524	11	20.4	17.9	11
89	12	2	12	3.1	2.0	12	1.6	1.3	11	15	13	11	11.0	1.6	12	102	8	12	6225	5739	12	7.9	5.9	12
90	16	8	12	2.8	1.4	11	1.6	0.9	12	14	9	12	11.3	1.7	12	102	16	12	8744	15112	11	8.6	8.6	11
91	13	7	12	2.6	1.4	12	1.4	1.0	12	12	8	12	12.1	1.5	12	109	12	11	2721	4165	11	7.0	5.4	11
92	12	3	12	2.5	1.6	12	1.1	0.6	11	10	5	11	11.0	1.7	12	106	13	12	3008	2375	12	8.4	7.0	12
93	13	3	12	3.5	1.1	12	2.1	0.9	11	18	6	11	11.6	1.9	12	107	13	12	3291	2105	11	9.4	5.6	12
94	12	2	12	3.1	1.3	12	1.5	1.0	12	15	10	12	10.8	1.4	12	101	5	12	4725	3439	12	12.6	6.3	12
95	17	9	12	2.3	1.2	12	0.9	0.7	12	8	6	12	11.2	1.7	12	103	11	12	7817	9872	12	17.3	15.1	12
Σ			244			228			238			236			247			239			201			199

Appendix II. Statistics for 16 variables sampled in the River Danube, based on calculated mean values for the period 1957 to 1995, where measurements are combined into months of the year (mon 1 = January to mon 12 = December). For abbreviations see Table 2; avg = arithmetic mean, std = standard deviation, n = number of observations without outliers, CV = coefficient of variance, CI = 95 %-confidence interval, Σ = overall statistic.

	Q (m^3/s)					**T (°C)**					**Cl (mg/l)**					**NO_3 (mg N/l)**				
mon	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI
1	1566	725	17	46	435	2.1	1.7	17	82	1.0	20	3	17	14	2	3.119	0.529	17	17	0.317
2	1798	634	18	35	367	3.2	1.4	18	44	0.8	21	2	18	11	1	3.358	0.383	17	11	0.230
3	1566	364	20	23	198	5.2	1.7	20	33	0.9	20	4	19	22	2	3.025	0.775	19	26	0.435
4	2246	796	26	35	372	9.4	1.6	25	17	0.8	16	3	24	16	1	2.342	0.575	24	25	0.281
5	2508	459	23	18	230	12.7	1.9	23	15	1.0	15	2	23	15	1	1.968	0.449	23	23	0.225
6	2547	710	17	28	426	15.2	1.6	18	11	0.9	12	2	18	12	1	1.683	0.466	18	28	0.270
7	2361	559	21	24	296	17.5	2.2	23	12	1.1	11	2	20	13	1	1.457	0.336	23	23	0.169
8	2092	655	19	31	368	18.3	2.5	20	14	1.3	13	2	19	15	1	1.581	0.264	19	17	0.148
9	1754	604	20	34	329	15.9	2.1	21	13	1.1	14	2	20	17	1	1.666	0.478	21	29	0.253
10	1296	508	30	39	219	12.0	1.7	30	14	0.7	15	3	29	17	1	2.265	0.537	28	24	0.241
11	1239	299	20	24	162	7.1	1.2	20	17	0.7	18	2	18	13	1	2.628	0.392	18	15	0.227
12	1556	881	18	57	510	4.2	1.7	18	41	1.0	19	3	18	14	2	2.873	0.320	18	11	0.186
Σ	1870	751	249	40	108	10.6	5.7	253	53	0.8	16	4	243	25	1	2.287	0.774	245	34	0.112

	NO_2 (mg N/l)					**NH_4 (mg N/l)**					**SRP (mg P/l)**					**TP (mg P/l)**				
	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI
1	0.019	0.006	17	32	0.004	0.306	0.170	15	55	0.110	0.155	0.077	17	49	0.046	0.209	0.090	17	43	0.054
2	0.020	0.006	17	31	0.004	0.312	0.151	18	48	0.087	0.125	0.048	18	39	0.028	0.207	0.075	18	36	0.044
3	0.020	0.008	19	39	0.004	0.232	0.167	18	72	0.097	0.123	0.083	19	68	0.047	0.218	0.101	17	47	0.061
4	0.017	0.007	22	38	0.003	0.163	0.110	23	68	0.055	0.088	0.060	24	68	0.029	0.196	0.075	19	38	0.042
5	0.019	0.008	20	42	0.004	0.109	0.056	22	51	0.029	0.064	0.040	23	62	0.020	0.168	0.071	20	42	0.039
6	0.021	0.008	18	36	0.004	0.123	0.070	18	57	0.040	0.070	0.037	18	53	0.021	0.161	0.064	17	39	0.038
7	0.018	0.013	20	73	0.007	0.110	0.057	19	52	0.032	0.066	0.031	22	47	0.016	0.143	0.052	17	36	0.031
8	0.021	0.010	19	48	0.006	0.142	0.099	19	69	0.055	0.083	0.045	20	54	0.024	0.159	0.062	18	39	0.036
9	0.020	0.009	21	44	0.005	0.110	0.054	21	49	0.029	0.092	0.045	20	49	0.025	0.149	0.047	18	32	0.027
10	0.020	0.009	28	47	0.004	0.134	0.080	27	60	0.036	0.132	0.061	27	46	0.028	0.206	0.105	20	51	0.057
11	0.030	0.010	18	33	0.006	0.195	0.130	18	67	0.075	0.139	0.060	18	43	0.035	0.189	0.083	18	44	0.048
12	0.029	0.011	18	36	0.006	0.229	0.112	18	49	0.065	0.153	0.069	18	45	0.040	0.213	0.097	17	45	0.058
Σ	0.021	0.010	237	45	0.001	0.174	0.126	236	72	0.019	0.106	0.064	244	60	0.009	0.185	0.081	216	44	0.012

Appendix II (continued)

	$KMnO_4$ (mg/l)					**BOD5** (mg O_2/l)					**BOD2** (mg O_2/l)					**BOD2** (%)				
	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI
1	18.2	5.1	17	28	3.0	2.2	1.2	17	56	0.7	1.1	0.6	17	55	0.4	9	5	17	56	3
2	19.4	7.2	18	37	4.2	2.4	1.2	18	52	0.7	1.0	0.3	17	33	0.2	10	9	18	91	5
3	18.3	4.8	19	26	2.7	2.4	0.9	17	38	0.6	1.3	0.8	18	59	0.4	10	5	18	51	3
4	19.9	4.1	22	21	2.1	3.7	1.6	21	43	0.8	2.2	1.2	22	52	0.6	18	9	22	52	5
5	19.8	5.7	23	29	2.9	3.3	1.2	23	38	0.6	1.8	1.0	23	52	0.5	16	8	23	49	4
6	17.2	6.8	17	39	4.1	3.0	0.9	18	31	0.5	1.5	0.5	18	34	0.3	14	4	18	32	3
7	17.4	6.6	22	38	3.4	2.3	0.8	18	37	0.5	1.2	0.7	21	60	0.4	13	7	21	59	4
8	18.5	8.2	20	44	4.5	3.0	2.1	18	71	1.2	1.2	0.9	17	75	0.5	11	5	16	48	3
9	16.4	5.0	21	30	2.6	2.5	1.8	18	73	1.1	1.2	0.8	21	68	0.4	12	8	21	66	4
10	20.3	7.8	29	38	3.4	2.9	1.5	25	52	0.7	1.9	1.4	29	73	0.6	17	10	27	60	5
11	16.2	4.8	18	30	2.8	2.0	0.8	17	42	0.5	1.1	0.6	18	58	0.4	10	6	18	60	4
12	18.2	6.5	18	36	3.7	2.1	0.8	18	39	0.5	0.9	0.6	17	61	0.3	8	4	17	55	3
Σ	18.4	6.2	244	34	0.9	2.7	1.4	228	52	0.2	1.4	0.9	238	67	0.1	13	8	236	62	1

	O_2 (mg/l)					**O_2** (%)					**HB** (CFU/ml)					**FC** (CFU/ml)				
	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI	avg	std	n	CV	95%-CI
1	12.2	0.9	17	7	0.5	93	7	17	7	4	8731	14022	13	161	9956	14.4	11.2	14	78	7.6
2	12.5	0.8	18	6	0.4	98	6	18	6	4	7816	8681	14	111	5876	10.8	8.7	14	81	5.9
3	12.2	1.4	19	12	0.8	101	10	19	10	6	5453	10136	17	186	6079	9.6	7.1	17	74	4.3
4	12.3	1.4	23	12	0.7	110	14	19	12	8	3750	3177	21	85	1680	10.1	9.6	21	95	5.1
5	11.3	1.2	23	10	0.6	108	13	22	12	6	3681	3866	18	105	2240	9.4	6.6	19	70	3.7
6	10.5	1.1	18	11	0.7	109	11	18	10	7	4957	7671	14	155	5193	16.1	14.1	14	87	9.5
7	9.6	0.8	22	9	0.4	105	10	22	9	5	3388	2663	17	79	1597	14.9	8.8	17	59	5.3
8	9.0	1.0	20	11	0.5	100	11	20	11	6	3980	3980	15	100	2579	19.3	14.8	15	77	9.6
9	9.6	1.0	21	10	0.5	103	12	21	11	6	3860	2510	15	65	1627	19.3	14.6	15	76	9.5
10	9.6	0.9	30	9	0.4	93	10	27	10	4	5866	4870	27	83	2230	22.9	20.1	24	88	9.8
11	10.6	0.8	18	7	0.4	93	7	18	8	4	6675	6524	16	98	4061	16.1	12.8	15	79	8.3
12	11.4	1.5	18	13	0.9	92	12	18	13	7	6693	8303	14	124	5620	11.0	7.8	14	70	5.3
Σ	10.8	1.6	247	15	0.2	100	12	239	12	2	5294	6810	201	129	1088	14.6	12.8	199	88	2.1

Appendix III a. Trends related to time, models (1) – (3): lg = $\log_{10}$-transformation; R^2 = adjusted coefficient of determination; p = probability; $b_o \ldots b_4$ = estimated coefficients (lin = linear, quad = quadratic, sin = sine, cos = cosine – coefficient). For abbreviations see Table 2.

	Trend model (1)							
Variable Y	R^2	p	b_1(lin)	p	b_2(quad)	p	b_0	p
Temperature	0.000	0.483	-0.670	0.254	0.0037	0.267	37.865	0.105
Discharge	0.000	0.828	-3.810	0.962	0.0010	0.998	2177.228	0.493
Chloride (lg)	0.127	<0.001	0.073	<0.001	-0.0004	<0.001	-1.809	<0.001
Nitrate (lg)	0.222	<0.001	0.095	<0.001	-0.0005	<0.001	-3.750	<0.001
Nitrite (lg)	0.014	0.076	0.054	0.024	-0.0003	0.026	-3.907	<0.001
Ammonium (lg)	0.149	<0.001	0.203	<0.001	-0.0013	<0.001	-8.859	<0.001
SRP	0.365	<0.001	0.059	<0.001	-0.0004	<0.001	-2.143	<0.001
Total phosphorus (lg)	0.579	<0.001	0.349	<0.001	-0.0022	<0.001	-14.658	<0.001
BOD5 (lg)	0.001	0.347	-0.085	0.220	0.0005	0.210	3.912	0.186
$KMnO_4$ (lg)	0.421	<0.001	0.071	<0.001	-0.0005	<0.001	-1.063	0.035
BOD2 (lg)	0.000	0.558	0.033	0.324	-0.0002	0.342	-1.280	0.336
BOD2 % (lg)	0.000	0.622	0.027	0.409	-0.0002	0.389	0.018	0.989
O_2 (lg)	0.086	<0.001	0.007	0.318	0.0000	0.529	0.651	0.016
O_2 % (lg)	0.069	<0.001	-0.003	0.590	0.0000	0.387	2.038	<0.001
HB (lg)	0.000	0.625	0.044	0.351	-0.0003	0.343	1.835	0.331
FC (lg)	0.000	0.445	-0.006	0.914	0.0000	0.986	1.469	0.511

	Sinusoid model (2)							
	R^2	p	b_3(sin)	p	b_4(cos)	p	b_0	p
Temperature	0.909	<0.001	-2.763	<0.001	-7.494	<0.001	10.182	<0.001
Discharge	0.313	<0.001	318.244	<0.001	-501.231	<0.001	1851.382	<0.001
Chloride (lg)	0.616	<0.001	0.038	<0.001	0.121	<0.001	1.199	<0.001
Nitrate (lg)	0.532	<0.001	0.058	<0.001	0.168	<0.001	0.341	<0.001
Nitrite (lg)	0.042	0.003	-0.034	0.073	0.059	0.004	-1.726	<0.001
Ammonium (lg)	0.223	<0.001	0.069	0.008	0.211	<0.001	-0.851	<0.001
SRP	0.249	<0.001	-0.012	0.020	0.045	<0.001	0.107	<0.001
Total phosphorus (lg)	0.053	0.001	0.024	0.224	0.069	0.001	-0.781	<0.001
BOD5 (lg)	0.075	<0.001	0.058	0.003	-0.064	0.001	0.374	<0.001
$KMnO_4$ (lg)	0.012	0.093	0.030	0.030	0.004	0.787	1.242	<0.001
BOD2 (lg)	0.055	<0.001	0.052	0.043	-0.088	0.001	0.073	<0.001
BOD2 % (lg)	0.090	<0.001	-0.003	0.900	-0.130	<0.001	1.040	<0.001
O_2 (lg)	0.569	<0.001	0.054	<0.001	0.042	<0.001	1.032	<0.001
O_2 % (lg)	0.284	<0.001	0.025	<0.001	-0.031	<0.001	2.000	<0.001
HB (lg)	0.049	0.003	-0.048	0.186	0.124	0.002	3.548	<0.001
FC (lg)	0.096	<0.001	-0.185	<0.001	-0.095	0.038	0.989	<0.001

	Combined model (3)											
	R^2	p	b_1(lin)	p	b_2(quad)	p	b_3(sin)	p	b_4(cos)	p	b_0	p
Temperature	0.913	<0.001	-0.287	0.098	0.0021	0.059	-7.529	<0.001	-2.771	<0.001	19.623	0.005
Discharge	0.307	<0.001	-15.135	0.819	0.0819	0.842	319.469	<0.001	-499.655	<0.001	2540.189	0.336
Chloride (lg)	0.726	<0.001	0.068	<0.001	-0.0004	<0.001	0.038	<0.001	0.119	<0.001	-1.554	<0.001
Nitrate (lg)	0.686	<0.001	0.082	<0.001	-0.0005	<0.001	0.053	<0.001	0.158	<0.001	-3.186	<0.001
Nitrite (lg)	0.056	<0.001	0.053	0.024	-0.0003	0.026	-0.036	0.060	0.058	0.004	-3.866	<0.001
Ammonium (lg)	0.361	<0.001	0.190	<0.001	-0.0012	0.000	0.064	0.006	0.206	<0.001	-8.261	<0.001
SRP	0.632	<0.001	0.057	<0.001	-0.0004	<0.001	-0.012	<0.001	0.046	<0.001	-2.074	<0.001
Total phosphorus (lg)	0.640	<0.001	0.344	<0.001	-0.0021	<0.001	0.017	0.152	0.072	<0.001	-14.444	<0.001
BOD5 (lg)	0.078	<0.001	-0.091	0.171	-0.0005	0.161	0.059	0.001	-0.064	0.003	4.171	0.142
$KMnO_4$ (lg)	0.443	<0.001	0.069	<0.001	-0.0005	<0.001	0.033	0.002	0.013	0.230	-0.961	0.052
BOD2 (lg)	0.052	0.003	0.032	0.323	-0.0002	0.345	0.049	0.056	-0.090	<0.001	-1.266	0.329
BOD2 % (lg)	0.086	<0.001	0.032	0.302	-0.0002	0.296	-0.005	0.854	-0.130	<0.001	-0.241	0.849
O_2 (lg)	0.616	<0.001	-0.001	0.888	0.0000	0.574	0.053	<0.001	0.403	<0.001	0.974	<0.001
O_2 % (lg)	0.364	<0.001	-0.004	0.445	0.0000	0.238	0.024	<0.001	-0.033	<0.001	2.054	<0.001
HB (lg)	0.045	0.011	0.047	0.306	-0.0003	0.293	-0.049	0.178	0.125	0.002	1.738	0.344
FC (lg)	0.094	<0.001	0.002	0.974	0.0000	0.901	-0.186	<0.001	-0.091	0.047	1.150	0.589

Appendix IIIb. Trends related to time, models (1) – (3): lg = $\log_{10}$-transformation; R^2 = adjusted coefficient of determination; p = probability; $b_o \ldots b_4$ = estimated coefficients (lin = linear, quad = quadratic, sin = sine, cos = cosine – coefficient). "Optimised" model is the final model with significant coefficients only (analysis with residuals when there is a trend!). For abbreviations see Table 2.

"Optimised" trend model (1)

Variable Y	R^2	p	b_1(lin)	p	b_2(quad)	p	b_0	p
Temperature	-							
Discharge	-							
Chloride (lg)	0.127	<0.001	0.073	<0.001	-0.0004	<0.001	-1.809	<0.001
Nitrate (lg)	0.222	<0.001	0.095	<0.001	-0.0005	<0.001	-3.750	<0.001
Nitrite (lg)	-							
Ammonium (lg)	0.149	<0.001	0.203	<0.001	-0.0013	<0.001	-8.859	<0.001
SRP	0.365	<0.001	0.059	<0.001	-0.0004	<0.001	-2.143	<0.001
Total phosphorus (lg)	0.579	<0.001	0.349	<0.001	-0.0022	<0.001	-14.658	<0.001
BOD5 (lg)	-							
$KMnO_4$ (lg)	0.421	<0.001	0.071	<0.001	-0.0005	<0.001	-1.063	0.035
BOD2 (lg)	-							
BOD2 % (lg)	-							
O_2 (lg)	0.088	<0.001	0.002	<0.001			0.818	<0.001
O_2 % (lg)	0.072	<0.001			0.0000	<0.001	1.917	<0.001
HB (lg)	-							
FC (lg)	-							

"Optimised" sinusoid model (2)

	R^2	p	b_3(sin)	p	b_4(cos)	p	b_0	p
Temperature	0.909	<0.001	-2.763	<0.001	-7.494	<0.001	10.182	<0.001
Discharge	0.313	<0.001	318.244	<0.001	-501.231	<0.001	1851.382	<0.001
Chloride (lg)	0.681	<0.001	0.038	<0.001	0.118	<0.001	0.005	0.152
Nitrate (lg)	0.587	<0.001	0.052	<0.001	0.155	<0.001	0.009	0.125
Nitrite (lg)	0.033	0.003			0.060	0.003	-1.725	<0.001
Ammonium (lg)	0.247	<0.001	0.063	0.008	0.204	<0.001	0.009	0.605
SRP	0.418	<0.001	-0.012	<0.001	0.045	<0.001	0.002	0.322
Total phosphorus (lg)	0.140	<0.001			0.071	<0.001	0.001	0.903
BOD5 (lg)	0.075	<0.001	0.058	0.003	-0.064	0.001	0.374	<0.001
$KMnO_4$ (lg)	0.036	0.002	0.032	0.002			-0.002	0.824
BOD2 (lg)	0.055	<0.001	0.052	0.043	-0.088	0.001	0.073	<0.001
BOD2 % (lg)	0.093	<0.001			-0.130	<0.001	1.040	<0.001
O_2 (lg)	0.573	<0.001	0.052	<0.001	0.040	<0.001	0.003	0.266
O_2 % (lg)	0.317	<0.001	0.024	<0.001	-0.033	<0.001	-0.001	0.715
HB (lg)	0.045	0.002			0.127	0.002	3.550	<0.001
FC (lg)	0.096	<0.001	-0.185	<0.001	-0.095	0.038	0.989	<0.001

"Optimised" combined model (3)

	R^2	p	b_1(lin)	p	b_2(quad)	p	b_3(sin)	p	b_4(cos)	p	b_0	p
Temperature	0.912	<0.001			0.0003	0.001	-2.785	<0.001	-7.536	<0.001	8.266	<0.001
Discharge	0.313	<0.001					318.244	<0.001	-501.231	<0.001	46.477	<0.001
Chloride (lg)	0.726	<0.001	0.068	<0.001	-0.0004	<0.001	0.038	<0.001	0.119	<0.001	-1.554	<0.001
Nitrate (lg)	0.686	<0.001	0.082	<0.001	-0.0005	<0.001	0.053	<0.001	0.158	<0.001	-3.186	<0.001
Nitrite (lg)	0.033	0.003							0.060	0.003	-1.725	<0.001
Ammonium (lg)	0.361	<0.001	0.190	<0.001	-0.0012	<0.001	0.064	0.006	0.206	<0.001	-8.261	<0.001
SRP	0.632	<0.001	0.057	<0.001	-0.0004	<0.001	-0.012	<0.001	0.046	<0.001	-2.074	<0.001
Total phosphorus (lg)	0.638	<0.001	0.346	<0.001	-0.0022	<0.001			0.071	<0.001	-14.543	<0.001
BOD5 (lg)	0.075	<0.001					0.058	0.003	-0.064	0.001	0.374	<0.001
$KMnO_4$ (lg)	0.442	<0.001	0.069	<0.001	-0.0005	<0.001	0.032	0.002			-0.994	0.045
BOD2 (lg)	0.055	0.001					0.052	0.043	-0.088	0.001	0.073	<0.001
BOD2 % (lg)	0.093	<0.001							-0.130	<0.001	1.040	<0.001
O_2 (lg)	0.618	<0.001			0.0000	<0.001	0.053	<0.001	0.040	<0.001	0.949	<0.001
O_2 % (lg)	0.366	<0.001			0.0000	<0.001	0.023	<0.001	-0.033	<0.001	1.912	<0.001
HB (lg)	0.045	0.002							0.127	0.002	3.550	<0.001
FC (lg)	0.096	<0.001					-0.185	<0.001	-0.095	0.038	0.989	<0.001

Appendix IV. Trends related to discharge (Q) and temperature (T), model (1): lg = $\log_{10}$-transformation; R^2 = adjusted coefficient of determination; p = probability; $b_0 \ldots b_2$ = estimated coefficients (lin = linear, quad = quadratic – coefficient). "Optimised" model is the final model with significant coefficients only. For abbreviations see Table 2.

	Discharge Q								Temperature T							
Variable Y	R^2	p	b_1(lin)	p	b_2(quad)	p	b_{01}	p	R^2	p	b_1(lin)	p	b_2(quad)	p	b_{01}	p
Chloride (lg)	0.204	<0.001	-0.0002	<0.001	2.60 E-08	0.004	1.420	<0.001	0.576	<0.001	-0.021	<0.001	0.0003	0.033	1.372	<0.001
Nitrate (lg)	0.144	<0.001	-0.0003	<0.001	5.55 E-08	<0.001	0.687	<0.001	0.507	<0.001	-0.026	<0.001	0.0002	0.342	0.570	<0.001
Nitrite (lg)	0.016	0.057	-0.0002	0.019	1.98 E-08	0.028	-1.515	<0.001	0.043	0.002	0.011	0.236	-0.0009	0.048	-1.721	<0.001
Ammonium (lg)	0.021	0.034	-0.0003	0.011	6.83 E-08	0.017	-0.525	<0.001	0.271	<0.001	-0.053	0.012	0.0012	0.038	-0.471	<0.001
SRP	0.132	<0.001	-0.0001	<0.001	1.30 E-08	0.019	0.215	<0.001	0.202	<0.001	-0.007	0.008	0.0001	0.471	0.165	<0.001
Total phosphorus (lg)	0.003	0.259	-0.0001	0.119	3.53 E-08	0.102	-0.649	<0.001	0.091	<0.001	0.001	0.878	-0.0006	0.166	-0.713	<0.001
$KMnO_4$ (lg)	0.072	<0.001	-0.0001	0.184	2.94 E-08	0.025	1.262	<0.001	0.069	<0.001	0.020	0.002	-0.0011	<0.001	1.193	<0.001
BOD5 (lg)	0.000	0.765	0.0000	0.983	2.87 E-09	0.888	0.359	<0.001	0.018	0.053	0.011	0.242	-0.0003	0.557	0.295	<0.001
BOD2 (lg)	0.000	0.894	-0.0001	0.643	1.15 E-08	0.666	0.130	0.275	0.033	0.007	0.024	0.055	-0.0007	0.223	-0.081	0.174
BOD2 % (lg)	0.000	0.961	0.0000	0.850	-6.14 E-09	0.817	1.030	<0.001	0.106	<0.001	0.032	0.009	-0.0007	0.194	0.815	<0.001
O_2 (lg)	0.031	0.009	-0.0001	0.003	1.78 E-08	0.002	1.103	<0.001	0.383	<0.001	-0.007	0.001	0.0000	0.825	1.104	<0.001
O_2 % (lg)	0.074	<0.001	0.0000	0.741	3.19 E-09	0.497	1.975	<0.001	0.937	<0.001	0.004	0.054	-0.0001	0.494	1.964	<0.001
HB (lg)	0.050	0.003	-0.0003	0.055	8.58 E-08	0.011	3.746	<0.001	0.034	0.013	0.004	0.844	-0.0008	0.340	3.622	<0.001
FC (lg)	0.000	0.568	-0.0002	0.360	4.15 E-08	0.310	1.140	<0.001	0.030	0.021	0.020	0.367	-0.0002	0.847	0.812	<0.001

	"Optimised" model (1) for Q								"Optimised" model (1) for T							
Variable Y	R^2	p	b_1(lin)	p	b_2(quad)	p	b_{01}	p	R^2	p	b_1(lin)	p	b_2(quad)	p	b_{01}	p
Chloride (lg)	0.204	<0.001	-0.0002	<0.001	2.60 E-08	0.004	1.420	<0.001	0.576	<0.001	-0.021	<0.001	0.0003	0.033	1.372	<0.001
Nitrate (lg)	0.144	<0.001	-0.0003	<0.001	5.55 E-08	<0.001	0.687	<0.001	0.507	<0.001	-0.021	<0.001			0.552	<0.001
Nitrite (lg)	-								0.042	0.001			-0.0004	0.001	-1.674	<0.001
Ammonium (lg)	0.021	0.034	-0.0003	0.011	6.83 E-08	0.017	-0.525	<0.001	0.271	<0.001	-0.053	0.012	0.0012	0.038	-0.471	<0.001
SRP	0.132	<0.001	-0.0001	0.001	1.30 E-08	0.019	0.215	<0.001	0.203	<0.001	-0.005	<0.001			0.159	<0.001
Total phosphorus (lg)	-								0.103	<0.001			-0.0005	<0.001	-0.707	<0.001
$KMnO_4$ (lg)	0.069	<0.001			1.24 E-08	<0.001	1.187	<0.001	0.069	<0.001	0.020	0.002	-0.0011	<0.001	1.193	<0.001
BOD5 (lg)	-								0.020	0.019	0.006	0.019			0.315	<0.001
BOD2 (lg)	-								0.031	0.004	0.009	0.004			-0.026	0.504
BOD2 % (lg)	-								0.103	<0.001	0.016	<0.001			0.872	<0.001
O_2 (lg)	-								0.385	<0.001	-0.007	<0.001			1.102	<0.001
O_2 % (lg)	0.078	<0.001			4.71 E-09	<0.001	1.981	<0.001	0.096	<0.001	0.003	<0.001			1.970	<0.001
HB (lg)	0.050	0.003	-0.0003	0.055	8.58 E-08	0.011	3.746	<0.001	0.039	0.003			-0.0006	0.003	3.637	<0.001
FC (lg)	-								0.034	0.005	0.016	0.005			0.827	<0.001

Appendix V. Trends related to time t, discharge Q and temperature T, combined model (4): lg = $\log_{10}$-transformation; R^2 = adjusted coefficient of determination; p = probability; $b_0 \ldots b_6$ = estimated coefficients (lin = linear, quad = quadratic, sin = sine, cos = cosine – coefficient). "Optimised" model is the final model with significant coefficients only. For abbreviations see Table 2.

	Simulation - model (4)															
Variable Y	R^2	p	b_1(lin)t	p	b_2(quad)t	p	b_3(lin)T	p	b_4(quad)T	p	b_5(lin)Q	p	b_6(quad)Q	p	b_{04}	p
Chloride (lg)	0.720	<0.001	0.0659	<0.001	-0.0004	<0.001	-0.015	<0.001	0.0001	0.376	-0.0001	0.006	7.01 E-09	0.205	-1.255	<0.001
Nitrate (lg)	0.689	<0.001	0.0789	<0.001	-0.0005	<0.001	-0.014	0.002	-0.0002	0.262	-0.0001	0.013	1.69 E-08	0.056	-2.798	<0.001
Nitrite (lg)	0.065	0.002	0.0569	0.017	-0.0003	0.020	0.016	0.115	-0.0010	0.028	-0.0001	0.098	3.17 E-08	0.116	-3.937	<0.001
Ammonium (lg)	0.421	<0.001	0.1761	<0.001	-0.0011	<0.001	-0.060	<0.001	0.0015	0.003	-0.0001	0.192	4.06 E-08	0.076	-7.162	<0.001
SRP	0.630	<0.001	0.0581	<0.001	-0.0004	<0.001	-0.006	0.001	0.0001	0.175	-0.0001	<0.001	1.38 E-08	<0.001	-1.955	<0.001
Total phosphorus (lg)	0.693	<0.001	0.3603	<0.001	-0.0022	<0.001	-0.006	0.245	-0.0001	0.693	-0.0003	<0.001	6.48 E-08	<0.001	-14.823	<0.001
$KMnO_4$ (lg)	0.598	<0.001	0.0703	<0.001	-0.0005	<0.001	0.001	0.806	-0.0003	0.161	-0.0001	0.001	4.41 E-08	<0.001	-0.927	0.031
BOD5 (lg)	0.007	0.270	-0.0706	0.315	0.0004	0.301	0.010	0.321	-0.0002	0.648	0.0000	0.716	8.18 E-09	0.697	3.260	0.278
BOD2 (lg)	0.037	0.023	0.0462	0.165	-0.0003	0.182	0.031	0.021	-0.0009	0.136	-0.0002	0.205	2.77 E-08	0.307	-1.884	0.163
BOD2 % (lg)	0.116	<0.001	0.0462	0.144	-0.0003	0.140	0.038	0.003	-0.0009	0.122	-0.0002	0.177	2.50 E-08	0.334	-0.872	0.496
O_2 (lg)	0.470	<0.001	-0.0001	0.979	0.0000	0.696	-0.007	0.002	0.0000	0.866	0.0000	0.905	2.72 E-09	0.540	0.982	<0.001
O_2 % (lg)	0.212	<0.001	0.0006	0.912	0.0000	0.798	0.004	0.044	-0.0001	0.367	0.0000	0.813	4.53 E-09	0.317	1.842	<0.001
HB (lg)	0.087	<0.001	0.0452	0.337	-0.0003	0.320	-0.012	0.517	-0.0002	0.852	-0.0002	0.303	6.29 E-08	0.065	2.030	0.280
FC (lg)	0.038	0.038	-0.0063	0.912	0.0000	0.998	0.013	0.564	0.0003	0.807	-0.0004	0.063	7.55 E-08	0.069	1.722	0.453

	"Optimised" simulation - model (4)															
Variable Y	R^2	p	b_1(lin)t	p	b_2(quad)t	p	b_3(lin)T	p	b_4(quad)T	p	b_5(lin)Q	p	b_6(quad)Q	p	b_{04}	p
Chloride (lg)	0.720	<0.001	0.0657	<0.001	-0.0004	<0.001	-0.013	<0.001			0.0000	<0.001			-1.292	<0.001
Nitrate (lg)	0.681	<0.001	0.0785	<0.001	-0.0004	<0.001	-0.018	<0.001			-0.0001	0.011	1.77 E-08	0.048	-2.755	<0.001
Nitrite (lg)	0.041	0.001							-0.0004	0.001					-1.674	<0.001
Ammonium (lg)	0.419	<0.001	0.1751	<0.001	-0.0011	<0.001	-0.060	<0.001	0.0015	0.003			1.16 E-08	0.024	-7.270	<0.001
SRP	0.629	<0.001	0.0584	<0.001	-0.0004	<0.001	-0.004	<0.001			-0.0001	<0.001	1.34 E-08	<0.001	-1.978	<0.001
Total phosphorus (lg)	0.693	<0.001	0.3593	<0.001	-0.0022	<0.001			-0.0004	<0.001	-0.0003	<0.001	6.48 E-08	<0.001	-14.804	<0.001
$KMnO_4$ (lg)	0.600	<0.001	0.0701	<0.001	-0.0005	<0.001			-0.0002	<0.001	-0.0001	0.001	4.41 E-08	<0.001	-0.917	0.032
BOD5 (lg)	0.020	0.021					0.006	0.021							0.314	<0.001
BOD2 (lg)	0.032	0.004					0.010	0.004							-0.027	0.498
BOD2 % (lg)	0.104	<0.001					0.016	<0.001							0.872	<0.001
O_2 (lg)	0.477	<0.001			0.0000	<0.001	-0.007	<0.001					3.26 E-09	<0.001	0.992	<0.001
O_2 % (lg)	0.219	<0.001			0.0000	<0.001	0.003	<0.001					3.66 E-09	<0.001	1.871	<0.001
HB (lg)	0.094	<0.001							-0.0008	<0.001			2.74 E-08	<0.001	3.548	<0.001
FC (lg)	0.035	<0.001					0.016	<0.001							12.337	<0.001