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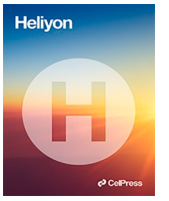

Exploring the trend effects of diffuse anthropogenic pollution in a large river passing through a densely populated area

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ABSTRACT

The detection of non-point pollution in large rivers requires high-frequency sampling over a longer period of time, which, however presumably provides data with large spatial and temporal variance. Variability may mean that data sets recorded upstream and downstream from a densely populated area overlap, suggesting at first glance that the urban area did not affect water quality. This study presents a simple way to explore trend-like effects of non-point pollution in the Danube based on data that varied strongly in space and time. For one year, biweekly sampling was carried out upstream and downstream from a large city with negligible emission of untreated wastewater and the surrounding settlements, industrial and agricultural areas.

Although most of the values of the 34 examined physicochemical characteristics fell within the range of data previously published for the Danube, and the mean values of all parameters indicated unpolluted surface water, different water quality was revealed upstream and downstream from the metropolitan area at each sampling time. Since the physicochemical characteristics causing the separation also differed from time to time, univariate tests and consensus ordination were used to determine which variables changed similarly during most of the examined period. With this evaluation method, several diffuse pollutants of anthropogenic origin contaminating the Danube in the long term were identified, such as nitrogen, phosphorus, sulphate, chloride, potassium and vanadium. The results demonstrated that trend-like effects of non-point pollution can be detected even in a large river, where physicochemical measurements can vary strongly in space and time.

1. Introduction

Anthropogenic pollution can reach rivers in various ways, in different amounts and at different levels of detectability. Point source pollution, such as untreated sewage input, enters the environment at an easily identifiable location and clearly and demonstrably alters water chemistry, increasing, for instance, electrical conductivity, alkalinity, the concentrations of heavy metals and trace elements [1–6]. Diffuse pollution, originated from agriculture and run-off from paved surfaces, among other sources, comes from many places and may have little impact individually, but together can significantly affect river chemistry. Of the macro elements, nitrate and phosphorus are reported as main nonpoint-source pollutants in rivers [7–10]. The extent of diffuse pollution is often difficult to determine. On one hand, the selection of the most suitable sampling locations in a long river section affected by diffuse pollution can be

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uncertain. On the other hand, changes in climatic conditions (temperature, precipitation), hydrological processes (droughts and floods), and human activities causing pollution can result in significant temporal variation in many physical and chemical parameters [11–14]. Therefore, the investigation of diffuse pollutants and their long-term effects in large rivers requires high-frequency sampling covering several seasons [11,14–18]. Furthermore, the extended and presumably highly variable datasets, that meet the above requirements, need reliable statistical analysis which can identify the main trends of water quality changes [11].

Densely populated areas can be both point and non-point sources of contaminants and where a river passes through those landscapes and drains large amounts of runoff water, sampling upstream and downstream from the urban area appears to be a suitable method for investigating both types of pollution [13,19–22]. Remarkably, studies focusing on anthropogenic impacts on urban rivers can detect the effects of point source urban wastewater input, but find little or no significant upstream and downstream differences in water quality when municipal wastewater treatment is effective, i.e. diffuse pollution becomes the main risk factor [9,23–25]. The reason for this could be that the investigated urban areas are not affected by significant diffuse pollution, or that the lack of sufficient data and/or the need for the appropriate mathematical evaluation of measurements extremely variable in space and time in previous studies concealed existing differences.

For the above reasons, the aim of this study is to present a simple method to evaluate high-frequency, spatially and temporally heterogenic measurements to reveal the trend-like effects of diffuse anthropogenic pollution on a large river.

2. Materials and methods

2.1. Sampling and in situ measurements

The study was carried out in the Danube river, in the area of Budapest, the capital of Hungary. Two study sites were designated in the main river channel (Fig. 1); one upstream and the other downstream from Budapest (between 1678–1674 and 1607–1604 river kms, respectively), therefore, the enclosed river section was not only affected by the pollution of the 525 km^2 metropolis with 1.7 million inhabitants, but also its agglomeration of numerous smaller settlements, as well as industrial and agricultural areas. Due to the installation of the central wastewater treatment plant in 2010, untreated wastewater discharge in Budapest has become negligible [26] and the water quality was found to be homogenous upstream and downstream from the metropolitan area [25].

During this study, sampling took place at six sampling points per study sites, every two weeks from February 2019 to February 2020, a total of 29 times. The six sampling points were located in three transects perpendicular to the shore, with two points per transect. The distance between the transects of a sampling site was 1-1,5 km. At each sampling time, the upstream samples were taken on the first day and the downstream samples were taken on the second day. Water discharge information was obtained from the Hungarian General Directorate of Water Management.

In situ physicochemical measurements were performed at each sampling point. Water temperature, pH and electrical conductivity were recorded by a Combo pH/EC/TDS/Temperature tester (HI 98129). Dissolved oxygen, redox potential and turbidity were measured by a portable dissolved oxygen meter (HI 9142), pH/Ion meter (WTW ProfiLine pH/ION 3310) and turbidity meter (Lovibond TB210), respectively.

For laboratory chemical analyses, water samples were collected by immersion from each sampling points. For the determination of the main ions and elements, the samples were collected in 0.5 L brown glass bottles without filtration. For the minor and trace elements, the samples were filtered directly after sampling through 0.45 μm pore size cellulose acetate syringe filters (VWR, Radnor, PA, USA) into 10 mL single use polypropylene centrifuge tubes and acidified with 100 μL 67% high-purity nitric acid (Normatom grade, VWR, Radnor, PA, United States). For the measurement of total organic carbon (TOC) content 30 mL glass bottles were used, and the water samples were acidified immediately with 200 μL of 2 M hydrochloric acid (diluted from 37% hydrochloric acid, a. r., Reanal Ltd., Hungary).

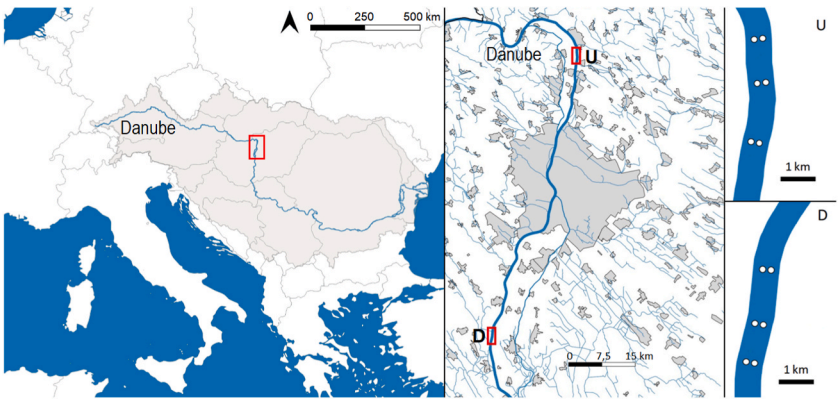


Fig. 1. Study sites designated in the Danube. Red frames indicate the enlargements. Left: The Danube watershed in Europe. Middle: Metropolitan area of Budapest with the upstream (U) and downstream (D) sample sites. Grey spots represent the metropolis and surrounding settlements, blue lines indicate watercourses. Right: Sample points in the upstream (U) and downstream (D) river sections.

2.2. Sample analysis

The concentration of the main cations and anions, as Na^+, Mg^{2+}, K^+, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} in the water samples were determined by ion chromatography (Dionex ICS 5000, Thermo Scientific, USA). Ortho-phosphate (PO_4^{3-}) and total phosphorus (TP) concentrations were determined by Spekord 210 Plus spectrophotometer (Analytik Jena, Germany), following Eaton et al. [27]. Total organic carbon (TOC), as well as total nitrogen (TN) concentrations, were determined by applying a Multi N/C 3100 TC-TN analyzer (Analytik Jena, Germany) equipped with a non-dispersive infrared detector and a chemiluminescent detector, in accordance with the corresponding international standards (MSZ EN 1484:1998, MSZ EN 12260:2004).

Minor and trace elements were determined using a Plasma Quant MS Elite inductively coupled plasma-mass spectrometer (ICP-MS, Analytik Jena, Germany) equipped with a reaction/collision cell in order to decrease spectral interferences. The operating conditions were published previously in detail by Dobosy et al. [28]. Analytical results were obtained on the basis of a single measurement of each sample consisting of 25 individual spectral scans. The limits of quantification (LOQ) was expressed as 10 sigma of the blanks.

2.3. Statistical analyses

Mann-Whitney-Wilcoxon test was used to reveal significant differences between the results of chemical measurements obtained upstream and downstream from the urban area. Differences with $p < 0.05$ were considered significant. Data from in situ measurements and laboratory chemical analyses were evaluated by standardized principal component analysis (PCA) for each sampling time (29 in total), using the SYN-TAX 2000 computer program package [29].

To obtain the consensus of the variables, Procrustes analysis was applied to the 29 PCAs. This method is a useful technique for comparing or synthesizing ordinations to evaluate the agreement between them and achieve the best fit, and can also be used to generate a consensus of more than two alternative ordinations [30].

3. Results

Water temperature, water flow and the basic descriptive statistical figures (minimum, maximum, mean and standard deviation) of physical and chemical measurements obtained from the upstream and downstream sites during the one-year study period are presented in Fig. 2 and Table 1, respectively. The water temperature ranged from 1.6 to 22.7 °C at the upstream and from 1.4 to 22.2 °C at the downstream river sections. Due to inflows entering the main Danube channel between the two study areas, water discharge was higher by 300–1500 m^3/s downstream than upstream from the urban area. Flows ranged between 1204-5028 and 763–3430 m^3/s in the former and the latter river sections, respectively. Three major flood events occurred during the study period; in March and May 2019 and in February 2020.

Most of the physical and chemical measurements showed high variance during the one-year study period with overlap between and upstream and downstream values (Table 1). The largest standard deviations were observed for turbidity, oxidation-reduction potential (ORP), and concentrations of total organic carbon, total phosphorus, Pb, Al, Fe, Mn, Cr, Cu, Cd, Zn, Mo and potassium ions. As a logarithmic measure, the narrow range of pH values represents considerable variability; the difference between the lowest and highest hydronium ion concentration is almost three orders of magnitude. Dissolved oxygen, electrical conductivity and the concentrations of Sr, Ba and sulphate changed the least.

Some physical and chemical characteristics showed a clear seasonal pattern (Suppl. Table 1). Water temperature unambiguously corresponded to the seasons, while dissolved oxygen varied inversely and reached the lowest values at the warmest months. Electrical

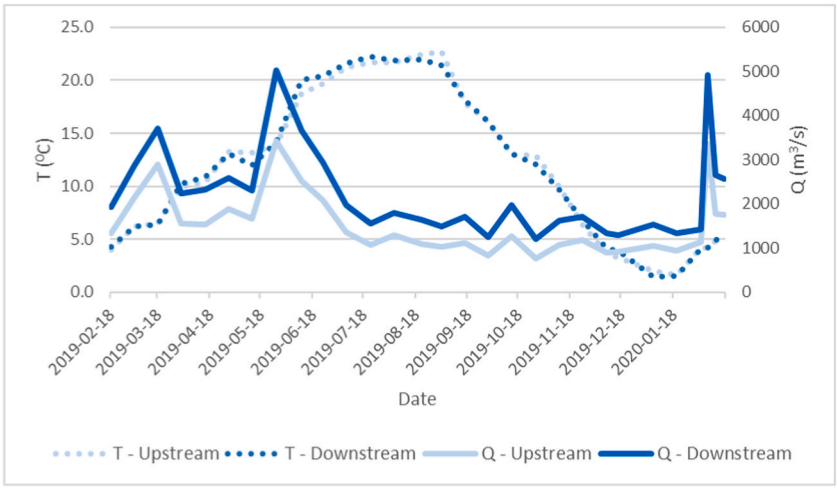


Fig. 2. Water temperature (T) and discharge (Q) at the upstream and downstream study sites during the sampling period.

Table 1
Basic descriptive statistical figures of physical and chemical measurements obtained from the upstream (U) and downstream (D) sites during the one-year study period.

	Min - Max		Mean ± SD	
	U	D	U	D
EC (μS/cm)	0.25–0.43	0.26–0.60	0.34 ± 0.05	0.35 ± 0.05
pH	6.04–8.90	7.08–8.35	7.64 ± 0.41	7.81 ± 0.32
Turb (NTU)	4.33–139	4.14–180	31.74 ± 34.15	28.80 ± 32.72
DO (mg/L)	7.40–12.40	7.50–12.50	9.90 ± 1.31	10.05 ± 1.31
Redox (mV)	5.00–269.70	3.80–232	97.31 ± 48.90	94.41 ± 39.84
TOC (mg/L)	1.14–6.12	1.08–6.24	2.08 ± 0.94	2.03 ± 0.96
TN (mg/L)	0.98–3.30	1.04–3.20	1.87 ± 0.61	1.91 ± 0.62
Na^{+} (mg/L)	3.37–25.04	4.68–22.05	11.98 ± 4.60	12.04 ± 4.52
Mg^{2+} (mg/L)	3.00–18.25	3.00–26.12	11.79 ± 3.29	12.68 ± 3.78
K^{+} (mg/L)	0.50–3.83	0.53–4.40	2.12 ± 0.92	2.18 ± 0.96
Ca^{2+} (mg/L)	33.62–89.14	28.84–85.86	56.65 ± 14.31	52.98 ± 13.17
Cl^{-} (mg/L)	10.22–39.82	10.45–33.90	18.76 ± 5.83	19.10 ± 5.65
$SO4^{2-}$ (mg/L)	17.71–37.06	18.58–38.43	27.32 ± 5.01	28.20 ± 5.21
NO^{3-} (mg/L)	3.47–11.12	3.48–11.24	7.13 ± 2.52	7.21 ± 2.48
TP (μg/L)	19.76–429.70	29.70–278.18	110.14 ± 49.89	116.55 ± 43.86
PO_4^{3-}-P (μg/L)	6.60–69.30	6.60–79.20	38.76 ± 12.84	47.04 ± 14.87
Fe (μg/L)	3.04–100.69	2.27–151.83	23.16 ± 18.96	25.41 ± 24.21
As (μg/L)	0.62–3.32	0.77–2.80	1.52 ± 0.53	1.73 ± 0.55
Se (μg/L)	0.10–0.53	0.10–0.49	0.17 ± 0.04	0.17 ± 0.05
B (μg/L)	10.31–58.32	11.91–42.70	19.36 ± 6.38	21.41 ± 6.10
Al (μg/L)	0.00–125.75	0.00–126.13	16.24 ± 19.65	18.22 ± 22.72
Sr (μg/L)	150.32–680.30	175.19–312.38	247.21 ± 47.06	245.09 ± 29.48
Mo (μg/L)	0.72–3.57	0.71–8.61	1.09 ± 0.32	1.14 ± 0.73
Cd (μg/L)	0.00–0.05	0.00–0.03	0.01 ± 0.00	0.01 ± 0.00
Sb (μg/L)	0.88–5.66	1.24–6.24	3.16 ± 0.86	3.27 ± 0.91
Ba (μg/L)	17.44–72.92	20.91–41.94	28.21 ± 5.20	28.80 ± 3.77
Pb (μg/L)	0.01–0.26	0.01–0.97	0.04 ± 0.04	0.05 ± 0.08
V (μg/L)	0.28–1.60	0.30–0.80	0.47 ± 0.14	0.48 ± 0.10
Cr (μg/L)	0.12–2.48	0.12–1.40	0.32 ± 0.29	0.30 ± 0.18
Mn (μg/L)	0.00–11.53	0.00–7.04	2.32 ± 2.05	1.89 ± 1.58
Co (μg/L)	0.01–0.10	0.01–0.09	0.05 ± 0.02	0.04 ± 0.02
Ni (μg/L)	0.56–3.53	0.55–2.73	1.00 ± 0.43	0.91 ± 0.25
Cu (μg/L)	0.50–7.19	0.50–6.10	1.22 ± 0.90	1.15 ± 0.60
Zn (μg/L)	1.18–15.96	1.28–13.89	4.18 ± 2.71	4.02 ± 1.86

EC – electric conductivity, Turb – turbidity, DO – dissolved oxygen, Redox – redox potential, TOC – total organic carbon, TN – total nitrogen. TP – total phosphorus. (n = 174).

conductivity and the concentrations of TN, NO_3^-, Cl^-, SO_4^{2-} showed the same tendency as dissolved oxygen. Arsenic reached highest concentration in August and September. Turbidity and TOC values peaked during flood events, when Al and Fe also reached their highest concentrations.

Multivariate statistical analysis of upstream and downstream water samples showed a clear spatial separation at all 29 sampling times, however, the variables causing the separation were different at time points. As an illustrative example, Fig. 3 shows the differences in downstream and upstream water samples taken on 8th July (Fig. 3A) and 14th October (Fig. 3B). Electrical conductivity,

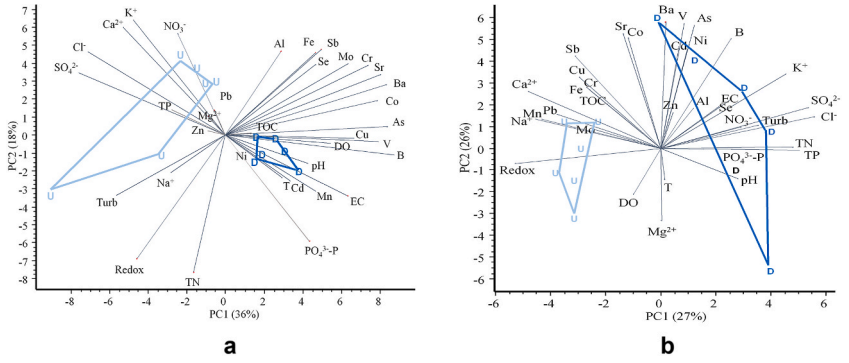


Fig. 3. PCA ordinations based on physicochemical characteristics of water samples taken on 8th July (a) and 14th October (b). Convex polygons enclose upstream (U) and downstream (D) samples. DO – dissolved oxygen, EC – electric conductivity, Redox – redox potential, T – water temperature, Turb – turbidity, TOC – total organic carbon, TP – total phosphorus, TN – total nitrogen.

pH and phosphate concentration were higher downstream than upstream at both sampling times, but the amount of sulphate, chloride, calcium, potassium and nitrate were higher upstream in July and downstream in October. Further differences were found when comparing the water quality of the two study sites at other times.

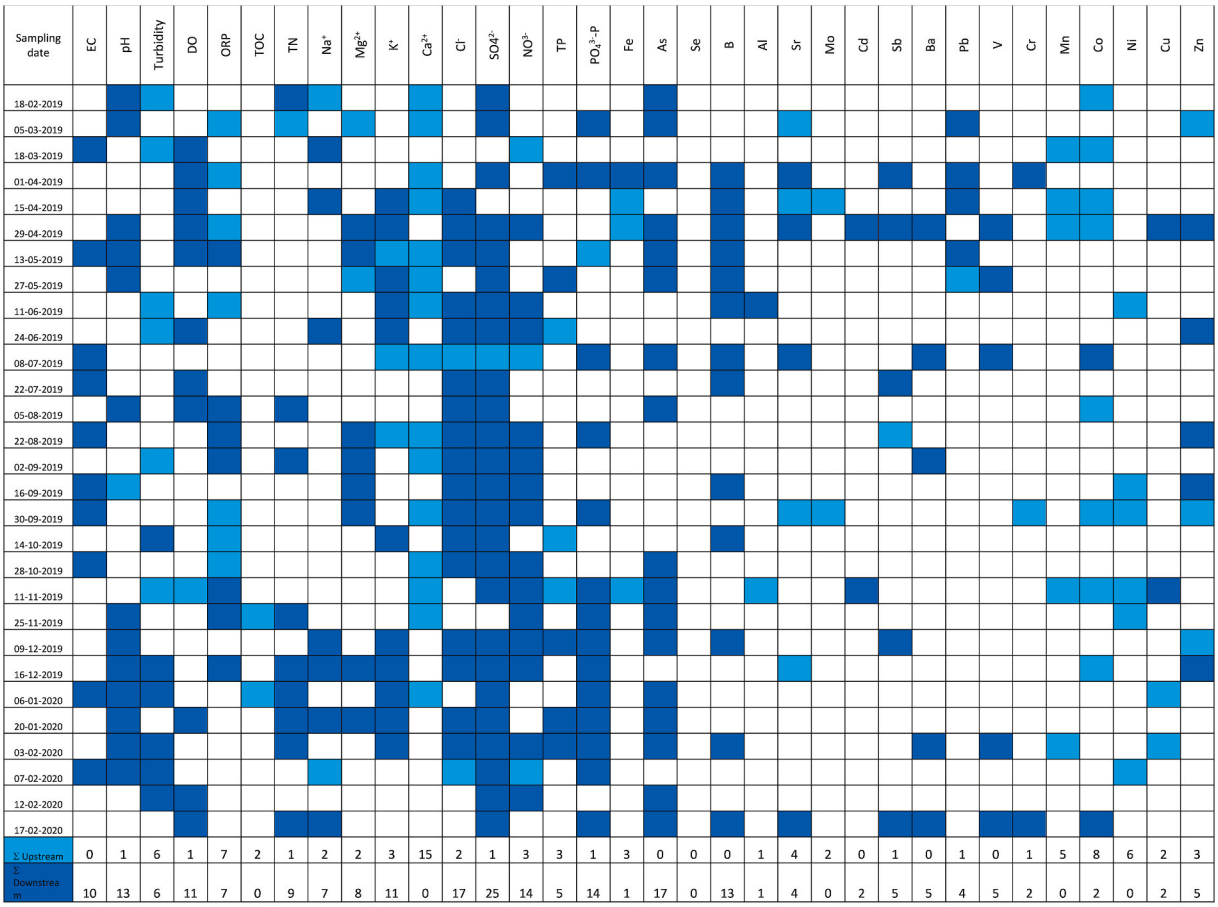
Table 2 provides a visual summary of significant differences between physical and chemical measurements of upstream and downstream water samples over the one-year study period. Mean values and standard deviation of all variables at each sampling time in both study locations are given in Suppl. Table 1. At almost all sampling times, i.e. 25 out of 29, the sulphate concentration was significantly higher downstream than upstream from the urban area. Further physicochemical variables that reached significantly greater values downstream than upstream at many sampling times were electric conductivity, pH, dissolved oxygen, total nitrogen, K^+, Cl^-, NO_3^-, PO_4^{3-}-P, As and B. Among the investigated variables, only Ca^{2+} concentration was significantly higher upstream than downstream in the majority of samplings and, less frequently, the concentrations of Co, Ni and Mn.

The consensus ordination of the ariables obtained from the PCA results of all sampling times revealed three main groups of the physicochemical measurements (Fig. 4). Calcium and Ba, V, B, As, SO_4^{2-}, PO_4^{3-}-P, K, total nitrogen, electric conductivity, pH, NO3-, Cl- and dissolved oxygen are at the two ends of the scattergram, while the rest of the variables form a group in the middle.

4. Discussion

Most of the physicochemical measurements recorded by this study fell within the range of data previously published for the Danube, or were close to it (cf. Table 3). However, the biweekly sampling over a year resulted in greater variability in some cases, with

Table 2
Visual representation of significant differences between upstream and downstream physicochemical measurements during the study period.



Light and dark blue indicate significantly higher values upstream and downstream, respectively, summed in the last two rows. DO – dissolved oxygen, EC – electric conductivity, Redox – redox potential, Turb – turbidity, TOC – total organic carbon, TP – total phosphorus, TN – total nitrogen. (Data are given in Suppl. Table 1.)

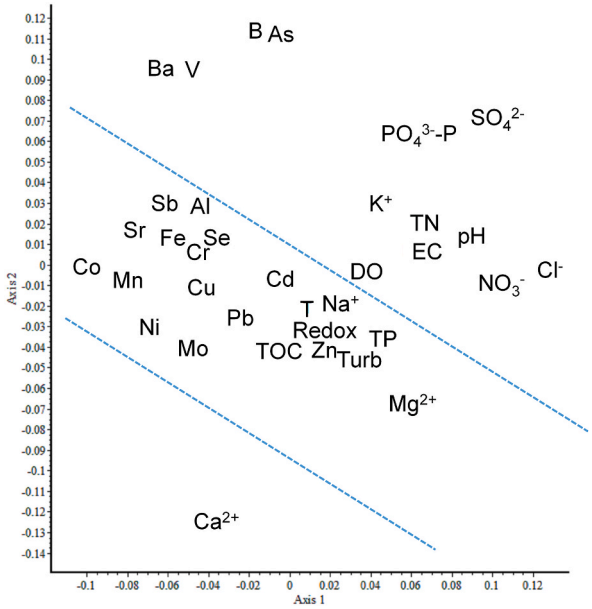


Fig. 4. The result of Procrustes analysis applied to the variables of all PCAs. The grouping of variables is indicated by dashed lines. DO – dissolved oxygen, EC – electric conductivity, Redox – redox potential, T – water temperature, Turb – turbidity, TOC – total organic carbon, TP – total phosphorus, TN – total nitrogen.

outlier minimum and/or maximum values (e.g. for turbidity, and the nitrate and phosphate concentrations). Nevertheless, the mean values for all parameters were in the range of unpolluted surface waters [31], and the maximum concentration data only slightly and temporarily rose above them. Only the concentration of Sb was found to be considerably higher than usual in freshwater [32] with a maximum value of 6.24 μg/L. In general, the highest concentrations recorded in many rivers of the world are in most cases higher than in the Danube (Table 3).

The reason for the significant variability of data obtained by the biweekly sampling is, on the one hand, that climatic and hydrological conditions, as well as the chemical and biological processes associated with them, caused temporal variability in many physicochemical parameters. Water temperature directly follows the air temperature [61] and the concentration of the dissolved oxygen changes reversely to the water temperature [11,62]. Seasonal fluctuations of NO_3^- concentration, i.e. it decreases in spring and increases in cold seasons, can be attributed to the consumption by phytoplankton and the reduction by denitrifying bacteria (high in spring, low in autumn and winter [53]). Our results also showed that turbidity reached its highest values at higher water discharges (flood events) and hydrological events resulted in higher TOC concentrations, as well. Turbidity can arise from organic or inorganic substances or a combination of both [14]. The typically particle-bound elements, as Al and Fe had also higher concentrations in the filtered samples during floods. Although the dissolved fraction (<0.2 μm particles) was analyzed for trace elements, the considerably elevated concentration of Fe and Al during flood events can be explained with the higher particle matter concentration, which can be seen on the turbidity values. In the presence of higher amount of organic matter – the TOC has its maxima at floods, too – these two elements tend to form stabile colloids with hydroxycarboxilic acids (e.g fulvic acid) and can appear in the "dissolved" fraction [63,64].

The annual variability of the measurements also meant that data series recorded at the two selected study sites were overlapped for all parameters, which at first glance may suggest that urban area did not affect water quality. This picture is a consequence of the fact that not only climatic and hydrologic conditions change over time, but also human activities that cause pollution in the metropolitan area. Analysing the data separately for each sampling time, the physicochemical composition of the upstream and downstream river water clearly differed, but the variables causing the separation also changed over time. The two procedures (i.e. the univariate tests and the consensus ordination of PCA's with Procrustes analysis) used to determine which variables changed similarly during most of the examined period yielded the same results. Thus, a number of physicochemical parameters were found (for instance, nitrogen, phosphorus, sulphate, chloride, electrical conductivity, potassium, boron and vanadium), the values of which increased in a trend-like manner in the water as the river passed through the densely populated area. These changes in water quality can be explained by diffuse anthropogenic pollution.

Although about 95% of the sewage water of Budapest is treated effectively, there are several other settlements and agricultural activities between the two sampling sites, which can be sources of nutrients like nitrogen and phosphorus compounds, all known as main nonpoint-source pollutants in rivers [7,8,10]. Typical urban origin can have chloride, alkaline metal ions and sulphate [65–67], affecting the electrical conductivity of the water body, an important parameter for determining the water quality for drinking and agricultural purposes [5]. Sulphate and potassium are used in fertilizers and can enter the river through runoff from croplands, potassium can be also enriched as a result of industrial and municipal pollution [65,67,68]. The source of boron can be the agricultural irrigation, since the ground waters in Hungary (also in the region of Budapest) can contain very high concentration, up to 10 mg/L

Table 3
Physico-chemical measurements from rivers worldwide (min-max or mean, rounded values).

	Europe						Asia			Africa	N-America	S-America	
	Danube	Seine	Orge	Thame	Po	Terme	Amur	Yangtze	Han	Nile	Mississippi	Orinoco	Rio Grande
EC	0.3–0.6					0.1–0.4		0.4–0.8		0.2–1.0		0.02–0.1	
pH	6.6–8.8	7.6–8.5		7.5–8.6		7.9–9.5	6.1–8.0	8.4–8.5		7.1–9.0	7.5–8.4	4.7–7.6	8.1–8.7
Turb	35–193	4.5–488				1.7–38.5		681-1000<		7.3–11.2			
DO	5.6–13.0					7.2–12.1				3.0–13.2			
Redox						−139–44.5							
TOC	2.7–3.3									4.59–6.65			
TN	0.9–4.9												
Na^{+}	6.0–33.0	1.9–13.7	13.1–33.7	12.6–71.3	5.2–22.0	5.6–48.4		34.9–87.3		4.0–72.8		0.7–2.5	24.0–47.9
Mg^{2+}	8.5–23.0	0.9–9.4	4.9–11.7	3.7	5.4–13.8	3.1–331		10.5–19.7		6.1–45.0	17.2–17.2	0.5–1.4	20.3–62.6
K^{+}	7.0–14.0	0.7–4.8	2.2–7.2	4.8	2.0–3.4	0.5–82.0		1.8–3.4		3.7–12.1		0.5–1.4	3.6–6.7
Ca^{2+}	20.0–81.0	74.0–7.6	18.2–141	80.0–154	35.2–69.4	11.3–159		37.0–53.7		9.4–41.2		1.9–5.9	28.2–51.7
Cl^{-}	8.0–39.0	3.8–38.4	19.0–56.0	18.0–112		0.0–3.1		55.5–156.1		6.2–96.8		0.1–1.9	9.9–29.6
$SO4^{2-}$	18.0–37.0	8.9–56.9	21.6–104	34.0–102.0		1.0–35.0		44.0–103		3.8–59.6		0.9–7.2	2.2–5.3
NO^{3-}	0.1–3.6	1.4–33.5	5.7–21.4	6.0–57.0				0.4–1.2		0.0–4.6		0.0–1.0	
TP	10.0–1050					0.0–1.8				15.0–998			
PO_4^{3-}-P	44.0–64.0	190–700								4.0–383			
Fe	15.6–560		12.0–79.0	12.0–3621	0.1–11.6	1.4–153.0	15.6–1064	1.7–37.7	0.6–85.6	7.7–2497	15.9–20.5	37.0–312.0	
As	0.5–4.3	0.4–1.3			0.6–2.1	0.3–1.0	0.2–2.4	10.5013.7	3.8–28.8				
Se							0.0–0.2		1.3–34.3				
B		36.3–87.0		68.0–565				66.3					
Al	0.3–0.4			0.0–3214	<30	2.1–337	1.9–139.0	9.1–31.8	8.6–520.4	1.7–113.6	18.2–24.2	27.0–305	
Sr		360–710	169–1650	335–975			40.3–130	755	155–320		100–102		168–334
Mo		0.0–2.2		0.0–7.0			0.0–1.4	0.8–1.7			1.1–1.9		0.7–1.9
Cd	0.0–0.4	0.0–2.4		0.0–1.0	0.0–0.6	0.7–1.0	0.0–1.8		0.3–5.9	0.1–8.	0.60		0.10
Sb			0.1–0.6				0.2–0.9	3.40	2.0–233				
Ba		23.7–30.7	24.0–56.0	18.0–61.0	17.0–37.0		5.0–28.0	46.90	43.7–161		50.6–73.1		30.5–44.6
Pb	0.1–3.8	0.1–1.0	0.0–0.4		0.1–1.2	0.3–2.2	0.0–1.3		2.3–26.1	5.0–61.6	0.0–0.3		
V			0.6–1.8	0.0–7.0			0.0–0.9		0.9–140		0.9–3.5		
Cr	0.2–4.0		0.1–1.2		0.4–2.2	0.8–1.3	0.0–1.9		6.7–20.4		0.2–0.3	0.1–0.6	
Mn	1.4–30.0	0.5–14.4	0.3–140	3.0–42.0	0.5–17.6	1.0–4.8	0.4–584	0.3–6.2	15.3–85.0	8.3–298	0.4–17.6	0.5–11.6	0.2–4.1
Co			0.1–0.4		0.0–0.2	1.2–1.3	0.0–1.2		0.4–6.2		0.50		
Ni	0.3–25.8		0.5–2.3	1.0–15.0	0.4–4.3	1.4–3.5	0.1–2.9		0.1–6.1	0.9–33.1	1.5–1.9		
Cu	1.1–11.8	0.5–3.5	0.5–2.2	0.0–14.0	1.0–3.0	1.2–3.7	0.9–17.6		0.7–46.4	3.9–51.3	1.7–2.5	1.0–5.5	
Zn	0.4–79.0		0.5–16.0	0.0–344	0.5–46.5	4.0–115	0.1–37.9	2.0–10.1		5.4–115	0.2–3.0	0.9–30.8	0.5–0.9
Refs	[11,24,33–42]	[12, 43–45]	[1]	[46]	[47]	[14]	[48,49]	[50,51]	[52]	[2,53,54,55]	[56,57]	[58,59]	[60]

The abbreviations and units of parameters are the same as in Table 1.

[69]. Groundwater can also contribute to the increase in arsenic concentration, as it can reach up to 87.5 μg/L in well water [69]. Nevertheless, the downstream concentrations of boron and arsenic is still far below the drinking water limit of 1.5 mg/L and 10 μg/L limit, respectively [70].

Sodium, vanadium and antimony often reached higher amounts in the downstream than the upstream water samples, which can also be considered anthropogenic. The sources of Na can be industrial effluent and domestic wastewater discharge [66,68,71]. Vanadium was previously characteristic for the region south of Budapest, because of the oil-fired power plant in Százhalombatta. This plant was switched to gas after 2000, however, the deposited V in the topsoil of the region is considerable [72]. Antimony has two main anthropogenic sources in the environment: brake pads of cars [73] and polyethylene terephthalate (PET) bottles, which readily emit Sb into the beverages contained in them [74]. In the latter case, Sb is excreted in urine and discharged through wastewater [75].

In contrast to the above parameters, calcium in surface water is primarily of natural origin, so changes in its concentration were more a function of natural hydro-meteorological variables than of man-made events [68].

In conclusion, although high-frequency sampling required to determine non-point pollution potentially provides data sets that are highly variable in time and space, proper evaluation of such data can reveal trend-like anthropogenic changes in water quality. Diffuse pollutants of agricultural, industrial and urban origin, such as nutrients, fertilizers, packaging material (PET) and transport contaminants can thus be detected in a large river flowing through a densely populated area. Of course, the extension of the examined parameters to emerging pollutants (such as pharmaceuticals, new pesticides, anti-corrosive substances and industrial additives) is necessary to get a more detailed picture of the effects of anthropogenic pollution of large rivers and their variability in space and time. The data evaluation method presented in this study can also be applied to those chemical characteristics.

Author contribution statement

Attila I. Engloner: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Kitti Németh: Performed the experiments; Analyzed and interpreted the data.

Péter Dobosy: Performed the experiments.

Mihály Óvári: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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