



1 Estimation of suspended loads in the Danube River at Göd (1668 river km), Hungary

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12 Abstract: Sediment rating curves were used to estimate suspended particulate matter  
13 (SPM) loads in the Danube River at Göd (1668 river km), Hungary, in conjunction with  
14 a sampling program conducted between 2003 – 2012. Contrary to its water quality  
15 significance, only a few studies have focused on the annual transport of SPM in this  
16 section of the river. Based on the results, we can state that 1) the SRC method (in  
17 certain cases with correction factors) provided reliable estimates of the annual SPM  
18 loads in this section of the river; 2) the division of the dataset into seasonal or  
19 temperature subsets did not significantly improve the estimations, moreover, annual  
20 datasets may provide additional hydrologic information on the water year or the annual  
21 water regime; 3) large amounts of the SPM were transported during short, but high

22 water discharge periods, hence, calendar based-sampling should be supplemented with  
23 event-based sampling, and 4) the SPM load of the river has declined by about 50% over  
24 previous decades, which is most likely due to the installation of hydropower plants on  
25 the upper (German, Austrian, Slovakian) stretches of the Danube River.

26  
27 **Keywords:** suspended particulate matter, sediment rating curve, Danube River, annual  
28 suspended particulate matter loads

## 29 30 1. Introduction

31 Suspended particulate matter (SPM) in streams and rivers is the solid fraction  
32 transported by the flow of water. SPM consists of inorganic (mainly silt and clay  
33 mineral grains, authigenic minerals), and organic particles (bacteria, phytoplankton,  
34 zooplankton, and plant and animal fragments, e.g., Schönborn, 1992). The concentration  
35 of SPM is controlled by a combination of water discharge and available particulate  
36 matter supply. SPM tends to settle under low flow conditions, and resuspends when  
37 flow increases. Hence, flow essentially determines the qualitative and quantitative  
38 properties of SPM, which is in a 'genetic' relationship with bed sediment (Oertel,  
39 1992). SPM causes turbidity and affects the spectral composition of the light penetrating  
40 a water body (Dvihally, 1979). SPM also plays a significant role in sorbing various  
41 inorganic (e.g., heavy metals) and organic (e.g., PCBs, PAHs) chemical constituents  
42 (e.g., Evans, et al., 1990; Oertel, 1994; Lin and Chen, 1998).

43 Studies focusing on SPM cover a wide variety of investigations including but not  
44 limited to: 1) determinations of the relationship between water discharge and SPM  
45 concentration and/or load and the spatial and temporal changes in this relationship (e.g.,  
46 Asselman, 2000); 2) evaluations of the chemical composition of SPM (e.g., Viers et al.,  
47 2009); 3) estimations of the annual loads of SPM and sediment-associated chemical  
48 constituents (Horowitz, 2010); 4) evaluating the effects of anthropogenic impacts (e.g.,  
49 hydropower dams; Klaver, et al., 2007); 5) calculations of the *effective discharge*  
50 (Wolman and Miller 1960); and 6) investigations of particulate organic matter  
51 (Reschke, et al., 2002). Since the amount of sediment delivered by a river contributes to  
52 its channel and landscape forming power (i.e., forming depositional zones, erosional  
53 zones, deltas), geomorphological processes also can be predicted based on the amount  
54 of SPM transported by streams and rivers (Syvitski, et al., 2005).

55 Many fluvial studies are designed to determine the concentration and load of SPM.  
56 Such programs require SPM sampling over a wide range of water discharge. However,  
57 most programs lack the resources to collect a sufficient number of samples to accurately  
58 estimate the annual SPM load. In the absence of actual samples, SPM concentrations  
59 can be estimated using sediment rating curves (SRCs) developed using log transformed  
60 data for SPM and Q; these curves can take the form of a power function:

$$61 \quad c = bQ^a \quad (1)$$

62 or, a linear equation:

$$63 \quad \log(c) = \log(b) + a \cdot \log Q \quad (2)$$

64 where:  $c$  is SPM concentration ( $\text{mg L}^{-1}$ ),  $Q$  is water discharge ( $\text{m}^3 \text{s}^{-1}$ ), and 'a' and 'b'  
65 are regression coefficients. This approach is widely used in studies focusing on the  
66 determination of SPM concentrations and annual loads (Achite and Ouillon, 2007;  
67 Horowitz, 2008; Gao and Josefson, 2012). Horowitz (2003) demonstrated that in certain  
68 cases, a second or a third order polynomial regression may provide more accurate  
69 estimates.

70 Certain corrections may be necessary when applying this approach. When the regression  
71 is fitted to log-transformed data, back-transformation to arithmetic space may cause a  
72 marked underestimation. To eliminate this bias, various correction factors may be  
73 required (e.g., Bradu and Mundlak, 1970; Duan, 1983; Ferguson, 1986). A second issue  
74 is associated with the degree of scatter in the SPM concentration vs. water discharge  
75 relationship. The reasons for this can be manifold. Hysteresis may lead to differences in  
76 the Q-SPM relationship for the falling and rising limbs of a hydrograph, (e.g., Williams,  
77 1989; Eder, et al., 2010). Seasonal differences (e.g. wet and dry periods) also can affect  
78 the accuracy of the regression (Asselman 2000). Further, the Q-SPM relationship can  
79 change from year-to-year, leading to differently shaped SRC-s, (convex, concave, or  
80 linear) as shown by Horowitz (2003) in a study of the Mississippi River (USA), or by  
81 Warrick, et al., (2013) in the study of northern California rivers.

82 Although relatively long-term datasets are available for the Hungarian section of the  
83 Danube River, where measurements usually have been collected on a weekly, or  
84 biweekly basis, there still is a need to estimate SPM concentrations for the unmeasured

85 periods to facilitate the determination of annual loads. The present study had three  
86 objectives. 1) to develop a reliable method for determining the relationship between Q  
87 and SPM concentration in the middle section of the Danube river; 2) to evaluate the  
88 current sampling strategy that has been applied for several years, to see if  
89 modification(s) are necessary; and 3) to develop a hydrological characterisation of the  
90 middle section of the river based on an evaluation of the available data and estimated  
91 annual loads. Although the Danube is the second longest river in Europe, only a few  
92 relatively recent publications focus on its sediment transporting characteristics within  
93 the Hungarian section. Baranya and Józsa (2013) evaluated an Acoustic Doppler  
94 Current Meter as a potential SPM concentration measuring tool. Others have  
95 investigated the contaminants associated with the suspended phases in the river, (e.g.,  
96 Andrási, et al., 2013; Faludi, et al., 2014). Long-term declines in SPM concentrations in  
97 the Hungarian section of the Danube River have been noted by Horváth and T. Bartalis,  
98 (1999); Tóth, et al., (2005) and Kiss, et al., (2007) but no actual load data were cited.  
99 The present study was designed to address this lack of SPM load data for the  
100 investigated section of the river, i.e. ~15 km up- and downstream from the gauging  
101 station (where there are no significant tributaries or anthropogenic impacts).

102

## 103 2. Materials and Methods

### 104 2.1. Study site and sampling method

105 The gauging station is located at Göd, at river km 1668 (distance from mouth). Göd is  
106 about 20 km upstream of Budapest, the capital of Hungary. The river catchment area at  
107 this site is 184 767 km<sup>2</sup> (Lászlóffy, 1965) (Fig. 1). The upper Danube River basin  
108 covers a large part of Southern Germany and the Austrian Alps. Vegetation is  
109 characterized by forests (40%), grasslands (27%), and arable land (23%). The texture of  
110 the soils in the area are silt loam and sandy loam, the soils in the mountainous areas  
111 range from clay to sand (Muerth, et al., 2010). The prevailing land use in the Danube  
112 River basin in Slovakia is agriculture (50%) and silviculture (43%) (www.icpdr.org,  
113 2014). The hydrologic characteristics of the river are basically determined by the size  
114 and the physiogeographic heterogeneity of the catchment area (the Alps, the North  
115 Carpathian Mountains), large-scale weather patterns, (Ludwig, et al., 2003),  
116 furthermore, anthropogenic impacts (numerous hydropower plants, river regulation)  
117 also are significant.

118 The average annual water discharge at the sampling site during the study period was 1  
119  $595 \pm 704 \text{ m}^3 \text{ s}^{-1}$  (avg.  $\pm$  stand. dev.), based on daily measurements (General Directorate  
120 of Water Management, 2011), and ranged between 580 and 5 820  $\text{m}^3 \text{ s}^{-1}$ . In addition,  
121 weekly water samples were collected between 2003 – 2011, close to the center line of  
122 the river. SPM concentrations were determined gravimetrically on the same day the  
123 samples were collected using filtration through pre-dried and pre-weighted 0.45- $\mu\text{m}$   
124 membrane filters (three replicates). Water temperature was measured *in situ*.

125

126 # Approx place of Figure 1. #

127

## 128 2.2. Rating Curve Development

129 As a first approach, the annual datasets of discharge and SPM concentration were log-  
130 transformed and then divided into validation and calibration subsets; the data were  
131 found to be normally distributed based on the Shapiro-Wilk test (Statistica 6.0  
132 Software). Linear, or second order polynomial curves were fitted, depending on the  
133 model efficiency. Model efficiency was evaluated by comparing model output (i.e.,  
134 values obtained using the calibration dataset) and the validation data. The criterion  
135 defined by Nash and Sutcliffe (1970) was used:

$$136 \quad NS = 1 - \frac{\sum_{i=1}^n (m_i - p_i)^2}{\sum_{i=1}^n (m_i - m_{avg})^2} \quad (3)$$

137 where:  $m_i$  is measured,  $p_i$  is the predicted concentration of SPM, and  $m_{avg}$  is the mean of  
138 the measured values. If the NS criterion is 1, it indicates perfect prediction, values lower  
139 than 0 show that using the average value for SPM provides better estimations than the  
140 model. To reduce the impact of extreme values (because of squared differences) the NS  
141 efficiency criterion was calculated using logarithmic values of the measured and  
142 predicted concentrations (Krause, et al., 2005).

143 Differences between predicted and observed concentrations were calculated, as follows:

$$144 \quad D(\%) = \frac{p_i - m_i}{m_i} \cdot 100 \quad (4)$$

145 where  $p_i$  is the predicted SPM concentration, and  $m_i$  is the measured SPM  
 146 concentration. Since differences can be both negative and positive, the absolute values  
 147 were used to characterize the effectiveness of the model. Correction factors described  
 148 by Bradu and Mundlak (1970), Duan (1983), and Ferguson (1986) also were tested, and  
 149 applied, when they improved the model (improved the NS criterion). Numerous  
 150 additional statistical measures can be applied, when evaluating model efficiency (e.g.  
 151 Hanna and Chang, 2012) e.g. fractional mean bias (FB), normalized mean-square error  
 152 (NMSE), geometric mean (MG), geometric variance (VG), fraction of predictions  
 153 within a factor of two of observations (FAC2), or the index of agreement (d) (e.g.  
 154 Krause, et al., 2005), however, none of these measures can generate perfect models. As  
 155 NMSE reflects both systematic and unsystematic errors (the lower the NMSE, the better  
 156 the prediction), and FAC2 is a robust measure (FAC2 should be in the range of 0.5–2)  
 157 not overly influenced by high or low outliers, these two measures were also used to  
 158 evaluate the models.

$$159 \quad NMSE = \frac{1}{N} \sum_{i=1}^n \frac{(p_i - m_i)^2}{\frac{1}{N} \sum_{i=1}^n p_i \cdot \frac{1}{N} \sum_{i=1}^n m_i} \quad (5)$$

$$160 \quad FAC2: 0.5 < \frac{p_i}{m_i} < 2 \quad (6)$$

161 where  $p_i$  is the predicted SPM concentration, and  $m_i$  is the measured SPM  
 162 concentration.

163 As an alternative, multiple regression analyses also were evaluated. The significant  
 164 predictors were chosen by both backward and forward stepwise regression (Statistica

165 6.0 Software). After this, the entire dataset (2003 – 2011) was divided into seasonal  
166 subsets based on the changes in daily water discharge (falling or rising limb of the  
167 hydrograph). The fitting and evaluative procedures that were used for the seasonal  
168 subsets were the same as previously described for the annual data sets.

169

### 170 3. Results

#### 171 3. 1. Annual SRCs

172 With the exception of 2011, every year of SPM data from the calibration subsets  
173 displayed normal distributions. Statistically, regression analysis should not be applied  
174 where the data are not normally distributed, however, despite this, the model for 2011  
175 appeared to work reasonably well (highest NS and highest  $r^2$  values, Table 1). Linear  
176 SRCs provided the best estimations for 2003, 2004, 2006, and 2008 whereas second  
177 order polynomial SRCs provided the best results for 2005, 2007, and 2009 – 2011 (Fig.  
178 2, Table 1.). Correction factors only improved the results in four cases; their values  
179 always were  $>1$ .

180 Average differences between predicted and measured values ranged between 26 and  
181 54%. It should be noted that when the SPM concentration range was  $\leq 5 \text{ mg L}^{-1}$ ,  
182 relatively small differences between the measured and predicted values, when expressed  
183 as a percentage, were quite high. However, these large errors are relatively insignificant  
184 in terms of estimating annual loads because the contributions for these periods also are  
185 relatively low. If the percentage errors in annual SPM load are excluded for those

186 periods when the concentrations were  $\leq 5 \text{ mg L}^{-1}$ , the range in estimation error declined  
187 to between 20–43%. NMSE values ranged between 0.12 and 0.34, and only 1–3 data  
188 pairs fell out of the range of FAC2, with the exception for 2003. The estimations were  
189 relatively inaccurate for 2003 based on the NS, NMSE, and FAC2 values; however, the  
190 average difference (D%) between predicted and measured values was not markedly  
191 high. There is a strong positive correlation ( $r=0.82$ ,  $p<0.05$ ) between the average annual  
192 SPM concentration (predicted data) and the average annual water yield, and there is a  
193 moderate positive correlation ( $r=0.52$ ,  $p<0.134$ ) between the average differences  
194 between predicted and measured SPM concentration ( $D_{\text{avg}}$ ) and the average annual  
195 water yield.

196

197 #Approx place of Table 1. #

198

199 #Approx place of Figure 2.#

200

201 Based on the annual SRCs that were developed, annual SPM load can be calculated  
202 using the following formula:

$$203 \text{ Annual SPM load (t)} = \sum_{i=1}^{365} c_i \cdot Q_i \cdot 86400 \cdot 10^{-6} \quad (7)$$

204 where:  $c_i$  is the SPM concentration ( $\text{mg L}^{-1}$ ),  $Q_i$  is the water discharge ( $\text{m}^3 \text{ s}^{-1}$ ), 86 400  
205 and  $10^{-6}$  are the necessary conversion factors to express the annual SPM load in tonnes

206 d<sup>-1</sup>. Table 2 contains annual estimates of the range of SPM concentration, the average  
207 SPM concentration, and the annual SPM loads.

208

209 #Approx place of Table 2. #

210

211 Based on a flow-duration curve for the study period (250 m<sup>3</sup> s<sup>-1</sup> flow classes) and the  
212 modelled interrelationship between discharge and SPM load, it is possible to determine  
213 the product of the instantaneous data. When the product reaches its maximum value, it  
214 identifies the discharge rate (*effective discharge*) that is responsible for the majority of  
215 the transported SPM, (Wolman and Miller, 1960; Sickingabula, 1999; Biedenharn, et  
216 al., 2000) (Figure 2.). For the Danube River, between 2003 – 2011, this value was 1750  
217 m<sup>3</sup> s<sup>-1</sup>, which is slightly higher than the average value determined for the study period  
218 (1595 m<sup>3</sup> s<sup>-1</sup>, based on daily measurements).

219

220 #Approx place of Figure 3.#

221

222 Cumulative plots of the SPM load (Figure 4.) show that ~40% of the total load was  
223 transported during periods when discharge was >2500 m<sup>3</sup> s<sup>-1</sup> (10% duration);  
224 furthermore, the river carried a significant amount of SPM (~12% total load for the  
225 period) when discharge >4000 m<sup>3</sup> s<sup>-1</sup>, even though it occurred on only 53 days during  
226 the 9-year long study period.

227

228 #Approx place of Figure 4.#

229

230 3.2. Results from the multiple regression analysis

231 Based on both stepup and stepdown regression modeling, two independent variables,  
232 (water discharge, water temperature) were significant predictors of SPM concentration.

233 No significant correlation was found between the two variables. Obviously, water  
234 temperature does not affect SPM concentration directly. However, it may reflect

235 biological activity and/or some seasonal characteristics. For example, the formation of  
236 biological detritus (which is a part of SPM; e.g. leaf breakdown rates) generally is

237 greater at higher temperature (e.g., Abelho, et al., 2005). On the other hand, heavy  
238 rainfall, which may cause substantial soil erosion in the catchment area, is more typical

239 during warmer seasons in the temperate zone. Statistically, the inclusion of this variable  
240 appears reasonable, based on the Akaike information criterion (AIC decreased from

241 240.3 to 219.0, and from 172.1 to 168.7 in the falling and rising limb, respectively.)

242 Using the multiple regression approach the average differences between predicted and  
243 measured values decreased slightly in the falling limb (from 38.6% to 38.3%) and

244 markedly in the rising limb (from 41.9% to 38.3% and 37.1 %). NMSE values  
245 decreased by 0.1 and 0.05 (falling and rising limb, respectively ), and less data pairs

246 were out of the range of the criterion FAC2 (falling limb: decreased by 3, rising limb:  
247 decreased by 4). (Table 3.).

248

249 # Approx place of Table. 3. #

250

251 3.3. Seasonal SRC-s and temperature classes

252 All the data in the seasonal subsets displayed normal distributions (falling and rising  
253 limbs were handled separately). Linear regression produced the best predictions in all  
254 the seasonal subsets of the falling limb and in the spring subset of the rising limb,  
255 second-order polynomial regression provided the best model in case of the winter,  
256 summer and autumn subsets of the rising limb. (Table 4.).

257

258 #Approx place of Table 4.#

259

260 Since seasonal water temperature data displayed an overlap (Fig. 5.), we created three  
261 temperature classes both for falling and for rising limbs and investigated whether this  
262 subdivision could produce better predictions.

263

264 #Approx place of Figure 5.#

265

266 Regression coefficients were similar in the T1 and T2 subsets (falling limb), and a  
267 second-order polynomial regression provided the best NS values in all the three

268 temperature subsets of the falling limb. The rising limb temperature subsets generated  
269 different shaped regression curves (second- order polynomials in the T2 and T3 subsets,  
270 linear regression in the T1 subset). Although relatively high NS values were associated  
271 with the T1 subset, accompanied by only small differences between predicted and  
272 measured data, it should be noted that the rising limb T1 subset did not contain a  
273 sufficient amount of validation data; hence, this result is tentative at best. Correction  
274 factors improved the model only in the two T3 subsets (Table 5.).

275

276 #Approx place of Table 5.#

277

#### 278 4. Discussion

279 In the Danube River, almost all the yearly datasets displayed normal distributions of the  
280 log-transformed data and the SRC method appeared to provide usable estimates of SPM  
281 concentration in the absence of actual samples for the study period (2003 – 2011). Even  
282 in the one case where the data were not normally distributed (2011), the SRC approach  
283 still appeared to work well. According to Horowitz (2003), differences between  
284 predicted and measured values that are within  $\pm 15 - 20\%$  fall within measurement error.  
285 On the other hand, Gray and Simões (2008) consider  $\pm 30 - 50\%$  an acceptable error.  
286 Our annual SRCs produced an error range of 20 – 42% with an average of 35%, our  
287 seasonal SRCs provided an error range of 25 – 51% with an average of 38%, and our  
288 temperature class SRCs provided an error range of 22 – 54% with an average of: 31%.

289 With the exception of 2003, an extremely dry year, all the NMSE and FAC2 measures  
290 showed reliable models. These various results are not markedly different from each  
291 other; however, they do exceed the differences attributable solely to measurement error.  
292 This may have resulted from the high variability in discharge and SPM concentrations  
293 that occurred during the study period.

294 In the Rhine River (near the German-Dutch border), Asselman (2000) demonstrated that  
295 the highest NS efficiency criteria can be obtained by splitting the entire dataset into  
296 seasonal subsets based on water level change (discharge), and applying a correction  
297 factor. However, Asselman's study did not investigate annual SRCs. Asselman's values  
298 (0.57 – 0.72) are higher than those for our seasonal predictions (0.24 – 0.61, average:  
299 0.40); however, the author also stated that NS values only can be compared for the same  
300 gauging stations. Hence, we can state that the NS criteria for our annual SRCs (0.24 –  
301 0.71, average: 0.49), or for our temperature class SRCs (0.25 – 0.78, average: 0.55)  
302 showed slightly, but not significantly better estimations than our seasonally generated  
303 SRCs (0.24 – 0.61, average: 0.41), based on the NS criterion. However, despite the NS  
304 criteria, and the differences between predicted and measured data, the division of the  
305 data into various subsets did not appear to bring about a significant improvement.  
306 Although in certain cases (e.g. rising limb, summer subset) all the model performance  
307 measures indicated very good estimations, the subdivision – with regard to the entire  
308 dataset – can not be considered either a simpler, or a better approach in our study. The  
309 multiple regression approach improved all the model efficiency measures in comparison

310 with the entire dataset (*cf.* table 1. all years, and table 3.), however, this subdivision  
311 does not appear to provide markedly better estimates, than the annuals SRCs. One of  
312 the potential benefits of using the annual SRC approach is that it may provide additional  
313 hydrologic information on the basis of the shapes of the annual curves: a convex curve  
314 indicates that the river probably is 'sediment-starved' (Horowitz 2003), which can be  
315 caused, for instance, by severe natural floods in the previous time interval and/or by  
316 various anthropogenic activities. Interestingly, the Danube River generated unusually  
317 high water levels in 2002, 2006, and 2010, but only the 2011 SRC displayed an obvious  
318 convex shape (Figure 2.). Based on that result, it appears that the Danube only became  
319 supply-limited in 2011 but not in 2002 nor 2006. Since floods can be caused by a  
320 variety of factors, and the catchment at Göd is quite extensive ( $>180.000 \text{ km}^2$ ) this kind  
321 of characterisation may not be possible at this time, but deserves some further  
322 investigation.

323 The annual rating curves for only 3 out of the 9 years within the study period showed  
324 improvement with the application of a correction factor to deal with the potential bias  
325 associated with converting from logarithmic space to arithmetic space. Further, there  
326 was no single valid correction factor. All the correction factors were positive, indicating  
327 a negative bias; however, based on this study, none of the applied correction factors  
328 could be considered more or less effective.

329 There was no obvious relationship between the applied model performance measures;  
330 however, in the case of the most inaccurate (e.g. year 2003, or falling limb, autumn) and

331 the most accurate (e.g. rising limb, summer) models, the measures were coherent with  
332 each other.

333

334 Based on the annual SRCs, annual SPM loads ranged between 0.72 – 2.2 Mt y<sup>-1</sup>  
335 (average = 1.6 Mt y<sup>-1</sup>) during the study period. These annual loads are on a par with the  
336 Dnieper (Russia) and Vistula (Poland) Rivers (Julien, 2002), the Rhine River (German-  
337 Dutch border; Asselman, 2000), the Seine River (France; Meybeck, et al., 2003), and  
338 the Lena River (Russia; Håkanson, et al., 2005).

339 In 1971, Bogárdi's monograph reported an average SPM concentration of 100 mg l<sup>-1</sup> for  
340 this section of the Danube between 1931 – 1940. In 1993, Rákóczi reported an annual  
341 load of 3.27 Mt for this section of the Danube River. Relative to the current results, it  
342 appears as if the annual SPM loads in the river have declined by about 50% since the  
343 1990s. The apparent reason for this decline is the installation of numerous hydropower  
344 dams in the upper stretches of the river. This rationale for the current decline in both  
345 SPM concentration and annual loads is in accord with similar findings elsewhere (e.g.,  
346 Syvitski, et al., 2005; Walling, 2006; 2008).

347 Our examination of the cumulative data for 2003 – 2011 clearly highlight the significant  
348 contribution of short-term, high flow events to the annual SPM loads in the Danube  
349 River. This once again tends to confirm the old adage that 90% of fluvial SPM is  
350 transported during 10% of the time (e.g., Horowitz, 2003). Hence, it would seem that  
351 event-based sampling, rather than calendar-based sampling (the current norm) probably

352 would provide more accurate estimations of annual SPM loads. However, the  
353 interrelationship between the accuracy of our estimated annual SPM loads and the  
354 annual water yield means that in wet years, the accuracy of the predictions might  
355 decline. Since climate change appears to manifest itself in more extreme weather  
356 phenomena (Andersen, et al., 2006; Steele-Dunne, et al., 2008; Guo, et al., 2014),  
357 annual weather and water yield data also should be taken into account when developing  
358 SRC models. The calculated effective discharge for this study is similar to the average  
359 measured discharge; and may indicate that these intervals may be hydrologically  
360 significant.

361

## 362 5. Conclusions

363 1) SPM concentrations and annual loads for the Danube River at Göd (1668 river km)  
364 during the study period could be estimated reasonably well using the annual SRC  
365 method; the annual estimates showed improvement in only a limited number of cases  
366 through the application of correction factors.

367 2) The division of the entire dataset into seasonal or temperature subsets did not  
368 markedly improve the accuracy of the annual SPM load estimates.

369 3) The calculated effective discharge of the Danube River at Göd was close to the  
370 average annual discharge; however, a significant amount of the annual loads of SPM  
371 occurred during short, but markedly elevated discharge events.

372 4) This implies that some improvement in the estimation of annual SPM loads could be  
373 achieved through the continued use of calendar-based sampling supplemented with  
374 specific event-based sampling.

375 5) Based on the results of this study, the SPM content of the Danube River has declined  
376 by close to 50% during the last two decades, probably as the result of the installation of  
377 numerous hydropower dams in the upper stretches of the basin.

378

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386

#### 387 References

- 388 Abelho, M., Cressa, C., Graça, M. A. S., 2005. Microbial biomass, respiration, and  
389 decomposition of *Hura crepitans* L. (Euphorbiaceae) leaves in a tropic stream.  
390 *Biotropica* 37, 397–402.
- 391 Achite, M., Ouillon, S., 2007. Suspended sediment transport in a semiarid watershed,  
392 Wadi Abd, Algeria (1973–1995). *J. Hydrol.* 343, 187–202.

393 Andersen, H. E., Kronvang, B., Larsen, S. E., Hoffman, C. C., Jensen, T. S.,  
394 Rasmussen, E. K., 2006. Climate-change impacts on hydrology and nutrients in a  
395 Danish lowland river basin. *Sci. Total Environ.* 365, 223–237.

396 Andrási, N., Molnár, B., Dobos, B., Vasanits-Zsigrai, A., Záray, Gy., Molnár-Perl, I.,  
397 2013. Determination of steroids in the dissolved and in the suspended phases of  
398 wastewater and Danube River samples by gas chromatography, tandem mass  
399 spectrometry. *Talanta* 115, 367–373.

400 Asselman, N. E. M., 2000. Fitting and interpretation of sediment rating curves. *J.*  
401 *Hydrol.* 234, 228–248.

402 Baranya, S., Józsa, J., 2013. Estimation of suspended sediment concentrations with  
403 ADCP in Danube River. *J. Hydrol. Hydromech.* 61, 232–240.

404 Biedenharn, D. S., Copeland, R. R., Thorne, C. T., Soar, P. J., Hey, R. D., Watson, C.  
405 C., 2000. Effective discharge calculation: A practical guide. Technical Report, U.S.  
406 Army Engineer Research and Development Center, Vicksburg MS. pp. 1–48.

407 Bogárdi, J., 1971. Sediment transport in alluvial streams (*in Hungarian*). Akad. Kiadó,  
408 Budapest. p. 837.

409 Bradu, D., Mundlak, Y., 1970. Estimation in Lognormal Linear Models. *J. Am. Stat.*  
410 *Assoc.* 65, 198–211.

411 Duan, N., 1983. Smearing estimate: A nonparametric retransformation method. *J. Am.*  
412 *Stat. Assoc.* 78, 605–610.

413 Dvihally, S. T., 1979. Trübung und selektive Lichtdurchlässigkeit des Donauwassers.  
414 Danub. Hung. XCI. Ann. Univ. Sci. Budapest., Sect. Biol. 20–21, 5–12.

415 Eder, A., Strauss, P., Krueger, T. and Quinton, J.N., 2010. Comparative calculation of  
416 suspended sediment loads with respect to hysteresis effects (in the Petzenkirchen  
417 catchment, Austria). *J. Hydrol.* 389, 168–176.

418 Evans, K. M., Gill, R. A., Robotham, P. W. J., 1990. The PAH and organic content of  
419 sediment particle size fractions. *Water Air Soil Poll.* 51, 13–31.

420 Faludi, T., Vasanits-Zsigrai, A., Záray, Gy., Molnár-Perl, I., 2014. Identification,  
421 quantification and distribution of substituted phenols in the dissolved and in the  
422 suspended phases of water samples by gas chromatography tandem mass spectrometry:  
423 Derivatization, mass fragmentation and acquisition studies. *Microchem. J.* *in press*,  
424 DOI: 10.1016/j.microc.2014.07.015.

425 Ferguson, R. I., 1986. River loads underestimated by rating curves. *Water Resour. Res.*  
426 22, 74–76.

427 Gao, P., Josefson, M., 2012. Temporal variations of suspended sediment transport in  
428 Oneida Creek watershed, central New York. *J. Hydrol.* 426–427, 17–27.

429 Gray, J. R., Simões, F. J. M., 2008. Estimating sediment discharge. *In: García, M. H.*  
430 *ed.: Sedimentation Engineering-Processes, Measurements, Modeling, and Practice,*  
431 *Manual.* pp. 1067–1088, American Society of Civil Engineers. Reston, Va, p. 1115.

432 Guo, B., Zhang, J., Gong, H., Cheng, X., 2014. Future climate change impacts on the  
433 ecohydrology of Guishui River Basin, China. *Ecohydrol. Hydrobiol.* 14, 55–67.

- 434 Håkanson, L., Mikrenska, M., Petrov, K., Foster, I., 2005. Suspended particulate matter  
435 SPM in rivers: empirical data and models. *Ecol. Model.* 183, 251–267.
- 436 Hanna, S., Chang, J., 2012: Acceptance criteria for urban dispersion model evaluation.  
437 *Meteorol. Atmos. Phys.* 116, 133–146.
- 438 Horowitz, A. J., 2003. An evaluation of sediment rating curves for estimating suspended  
439 concentrations for subsequent flux calculations. *Hydrol. Process.* 17, 3387–3409.
- 440 Horowitz, A. J., 2008. Determining annual suspended and sediment associated-trace  
441 element and nutrient fluxes. *Sci. Total Env.* 400, 315–343.
- 442 Horowitz, A. J., 2010. The use of instrumentally collected-composite samples to  
443 estimate the annual fluxes of suspended sediment and sediment-associated chemical  
444 constituents. *IAHS Publ.* 337, 273–281.
- 445 Horváth, L. T. Bartalis, É., 1999. Water chemical characterization of the Danube River  
446 between Rajka and Szob (*in Hungarian*). *Vízügyi Közl.* 1, 54–85.
- 447 Julien, P.-Y., 2002. *River Mechanics*. Cambridge, Cambridge University Press, p. 434.
- 448 Kiss, K. T., Ács, É., Szabó, K., 2007. Algae and material cycles (*in Hungarian*). *In.:*  
449 *Nosek, J. – Oertel, N. eds: „A Dunának, mely múlt, jelen s jövő...” 50 éves az MTA*  
450 *Magyar Dunakutató Állomás.* p. 33–51. Dandera Bt. Erdőkertes, p. 190.
- 451 Klaver, G., Bertil van Os, B., Negrel, P. and Petelet-Giraud, E., 2007. Influence of  
452 hydropower dams on the composition of the suspended and riverbank sediments in the  
453 Danube. *Environ. Pollut.* 148, 718–728.

454 Krause, P., Boyle, D. P., Bäse, F., 2005: Comparison of different efficiency criteria for  
455 hydrological model assessment. *Adv. Geosci.* 5, 89–97.

456 Lászlóffy, W., 1965. Die Hydrographie der Donau. *In: Liepolt, R. (ed.): Limnologie der*  
457 *Donau – Eine monographische Darstellung II: 16–57.* Schweizerbart, Stuttgart.

458 Lin, J.-G., Chen, S.-Y., 1998. The relationship between adsorption of heavy metal and  
459 organic matter in river sediments. *Environ. Int.* 24, 345–352.

460 Ludwig, R., Mauser, W., Niemeyer, S., Colgan, A., Stolz, R., Escher-Vetter, H., Kuhn,  
461 M., Reichstein, M., Tenhunen, J., Kraus, A., Ludwig, M., Barth, M., Hennicker, R.,  
462 2003: Web-based modelling of energy, water and matter fluxes to support decision  
463 making in mesoscale catchments - the integrative perspective of GLOWA-Danube.  
464 *Phys. Chem. Earth* 28, 621–634.

465 Nash, J. E., Sutcliffe, J. V., 1970. River flow forecasting through conceptual models,  
466 Part 1: A discussion of principles. *J. Hydrol.* 10, 282–290.

467 Meybeck, M., Laroche, L., Dürr, H. H., Syvitski, J. P. M., 2003. Global variability of  
468 daily Total Suspended Solids and their fluxes in rivers. *Glob. Planet. Chang.* 39, 65–93.

469 Muerth, M., Mauser, W., Heinzeller, C., 2010. Impact of potential climate change on  
470 plant available soil water and percolation in the Upper Danube basin. IAHS  
471 Publications, Hydropredict 2010 Conference Papers, Prague, Czech Republic, Proc. Nr.  
472 126.

473 Oertel, N., 1992. Heavy metals in the water, in the suspended matter and in the  
474 organisms of the periphyton. (*in Hungarian*). PhD theses.

475 Oertel, N., 1994. Trend analysis of heavy metal concentration of the suspended matter  
476 in the river Danube. *Water Sci. Technol.* 29, 141–143.

477 Rákóczi, L., 1993. Sediment regime of the Danube (*in Hungarian*). *Vízügyi*  
478 *közlemények* 75, 129–149.

479 Reschke, S., Ittekkot, V., Panin, N., 2002. The Nature of organic matter in the Danube  
480 River particles and North-western Black Sea sediments. *Estuar. Coast. Shelf Sci.* 54,  
481 563–574.

482 Schönborn, W., 1992. *Fliessgewässerbiologie*. Gustav Fischer Verlag Jena, Stuttgart. p  
483 504.

484 Sickingabula, H. M., 1999. Magnitude-frequency characteristics of effective discharge  
485 for suspended sediment transport, Fraser River, British Columbia, Canada. *Hydrol.*  
486 *Process.* 13, 1361–1380.

487 Steele-Dunne, S., Lynch, P., McGrath, R., Semmler, T., Wang, S., Hanafin, J., Nolan,  
488 P., 2008. The impacts of climate change on hydrology in Ireland. *J. Hydrol.* 356, 28–45.

489 Syvitski, J. P. M., Vörösmarty, C., Kettner, A. J., Green, P., 2005. Impact of Humans on  
490 the Flux of Terrestrial Sediment to the Global Coastal Ocean. *Science* 308, 376–380.

491 Tóth, B., Nosek, J., Oertel, N., 2005. Long-term changes of the organic matter and the  
492 suspended matter content of the River Danube (*in Hungarian*). *Hidrológiai Közlöny* 85:  
493 152–154.

494 Viers, J., Dupré, B. and Gaillardet, J., 2009. Chemical composition of suspended  
495 sediments in World Rivers: New insights from a new database. *Sci.Total Environ.* 407,  
496 853–868.

497 Walling, D. E., 2006. Human impact on land ocean sediment transfer by the world's  
498 rivers. *Geomorphology* 79, 192–216.

499 Walling, D. E., Collins, A. L., 2008. The catchment sediment budget as a management  
500 tool. *Environ. Sci. Policy* 11, 136–143.

501 Warrick, J. A., Madej, M. A., Goñi, M. A., Wheatcroft, R. A., 2013. Trends in the  
502 suspended-sediment yields of coastal rivers of northern California, 1955–2010. *J.*  
503 *Hydrol.* 489, 108–123.

504 Williams, G.P., 1989. Sediment concentration versus water discharge during single  
505 hydrologic events in rivers. *J. Hydrol.* 111, 89–106.

506 Wolman, M. G., Miller, J. P., 1960. Magnitude and frequency of forces in geomorphic  
507 processes. *J. Geol.* 68, 54–74.

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509 <http://icpdr.org/main/danube-basin/slovakia>

510 Table 1. Regression coefficients of rating curves for the Danube River at Göd (1668 river km), Hungary, between 2003  
 511 and 2011, and model efficiency measures.

Water year	$\log c = a \cdot \log Q + b$		$\log c = a \cdot (\log Q)^2 + b \cdot \log Q + d$			CF	NS	$\overline{ D }$ %	$\overline{ D_s }$ %	$r^2$	N <sub>Cal</sub>	N <sub>Val</sub>	NMSE	FAC2
	a*	b	a	b	d									
2003	1.0509	-2.0919				-	0.24	30.1	30.1	0.28	23	22	0.50	10
2004	1.3043	-2.9325				B-M	0.56	46.9	31.2	0.54	24	23	0.25	3
2005			-1.4408	10.194	-16.473	-	0.46	53.6	37.0	0.43	20	21	0.34	3
2006	1.1815	-2.4518				-	0.58	52.8	43.3	0.51	20	19	0.17	3
2007			2.9687	-18.305	29.504	B-M	0.24	45.4	45.4	0.31	20	17	0.14	2
2008	1.8295	-4.6784				-	0.65	25.8	19.9	0.48	21	20	0.12	2
2009			-1.2131	9.0378	-15.148	-	0.59	40.0	40.0	0.62	20	20	0.12	1
2010			-3.222	22.549	-37.78	1.212	0.37	37.7	37.7	0.30	20	14	0.24	2
2011			-13.474	85.414	-133.89	-	0.71	40.5	33.7	0.76	20	14	0.22	1
All years	1.4146	-3.2353				1.167	0.41	47.2	41.9	0.43	180	178	0.33	29

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 513 c: suspended particulate matter (SPM) concentration (mg L<sup>-1</sup>). Q: discharge (m<sup>3</sup> s<sup>-1</sup>). a, b, d: regression coeff. CF:  
 514 correction factor (B-M refers to Bradu Mundlak, individual value for all predicted SPM concentrations), NS: Nash-  
 515 Sutcliffe efficiency criterion, D: Difference between predicted and observed concentration. D<sub>s</sub>: Difference between

516 predicted and observed concentration for those periods when the SPM concentrations were  $\geq 5 \text{ mg L}^{-1}$ .  $r^2$ : coeff. of  
517 determination,  $N_{\text{Cal}}$ ,  $N_{\text{Val}}$ : number of data pairs in the calibration and validation subsets, respectively, NMSE: normalized  
518 mean-square error, FAC2: number of cases when  $p_i/m_i$  is out of the range of 0.5–2.

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523 Table 2. Annual suspended matter concentration ranges, average values and loads in the Danube River at Göd, Hungary  
524 (2003–2011).

525

	<b>min</b>	<b>max</b>	<b>average</b>	<b>St.dev.</b>	<b>cv%</b>	<b>Load</b>
	<b>(mg L<sup>-1</sup>)</b>					<b>(Mt year<sup>-1</sup>)</b>
2003	6.4	41.8	15.1	6.7	44.4	0.72
2004	6.7	42.4	17.7	7.8	44.1	0.96
2005	9.0	36.2	23.3	8.5	36.4	1.44
2006	8.9	99.2	25.8	17.9	69.2	2.07
2007	20.1	176.7	25.0	16.3	65.4	1.44
2008	4.3	47.5	14.4	7.8	53.9	0.79
2009	7.7	48.1	23.4	10.7	45.7	1.55
2010	7.7	56.6	32.8	14.5	44.2	2.20
2011	0.1	29.7	19.7	9.1	46.0	0.86

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529 Table 3. Regression coefficients of the sediment rating curves for the Danube River at Göd (1668 river km), Hungary,  
 530 between 2003–2011, and model efficiency measures.

Subset	$\log c = a \cdot (\log Q) + b \cdot T + d$			CF	NS	$\overline{D}$	$\overline{D}_s$	AIC	$r^2$	$r^2_{\log Q}$	$r^2_T$	N <sub>Cal</sub>	N <sub>Val</sub>	NMSE	FAC2
	a	b	d												
Fall	1.424	0	-3.2634	1.159	0.46	51.1	38.6	240.3	0.46			101	103	0.29	16
Rise	1.18	0	2.5086	1.175	0.39	56.8	45.5	172.1	0.36			77	77	0.30	15
Fall T	1.21	0.013	-2.715	-	0.56	44.1	38.3	219.0	0.55 0.45			101	103	0.19	13
Rise T	1.06	0.016	-2.33	1.139	0.53	42.8	37.1	168.7	0.51 0.36			77	77	0.25	11
									0.20						

531  
 532 Subsets: Fall, rise: falling and rising limb, only predictor is water discharge. Fall T, Rise T: falling and rising limbs,  
 533 predictors are water discharge and temperature. c: suspended particulate matter (SPM) concentration ( $\text{mg L}^{-1}$ ). Q:  
 534 discharge ( $\text{m}^3 \text{s}^{-1}$ ). T: water temperature ( $^{\circ}\text{C}$ ). a, b, d: regression coeff. CF: correction factor (B-M refers to Bradu  
 535 Mundlak, individual value for all predicted SPM concentrations), NS: Nash-Sutcliffe efficiency criterion, D: Difference  
 536 between predicted and observed concentration.  $D_s$ : Difference between predicted and observed concentration for those

537 periods when the SPM concentrations were  $\geq 5$  mg L<sup>-1</sup>. AIC: Akaike information criterion.  $r^2$ : coeff. of determination,  
538  $N_{\text{Cal}}$ ,  $N_{\text{Val}}$ : number of data pairs in the calibration and validation subsets, respectively, NMSE: normalized mean-square  
539 error, FAC2: number of cases when  $p_i/m_i$  is out of the range of 0.5–2.

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544 Table 4. Regression coefficients of the sediment rating curves for the Danube River at Göd (1668 river km), Hungary,  
 545 between 2003–2011, and model efficiency measures.

Subset	$\log c = a \cdot \log Q + b$		$\log c = a \cdot (\log Q)^2 + b \cdot \log Q + d$			CF	NS	$\overline{ D }$ %	$\overline{ D_s }$ %	$r^2$	$N_{\text{Cal}}$	$N_{\text{Val}}$	NMSE	FAC2	
	a	b	a	b	d										
Fall	Wi	1.7736	-4.4026												
	Sp	0.6367	-0.7403												
	Su	0.5926	0.4498												
	Aut	1.5436	-3.606												
Rise	Wi			2.143	-12.001	17.579	B-M	0.24	76.9	45.0	0.31	18	7	0.20	3
	Sp	1.0259	-1.9698					0.31	35.1	35.1	0.30	21	21	0.51	6
	Su			0.6919	-3.1741	4.4431	B-M	0.58	24.7	24.7	0.61	20	15	0.15	0
	Aut			2.9876	-16.793	24.443	1.098	0.61	61.8	31.6	0.60	19	12	0.17	0

546  
 547 Subsets: Wi: winter, Sp: spring, Su, summer, Aut: autumn. c: suspended particulate matter (SPM) concentration ( $\text{mg L}^{-1}$ ).  
 548 Q: discharge ( $\text{m}^3 \text{s}^{-1}$ ). a, b, d: regression coeff. CF: correction factor (B-M refers to Bradu Mundlak, individual value for  
 549 all predicted SPM concentrations), NS: Nash-Sutcliffe efficiency criterion, D: Difference between predicted and observed  
 550 concentration.  $D_s$ : Difference between predicted and observed concentration for those periods when the SPM  
 551 concentrations were  $\geq 5 \text{ mg L}^{-1}$ .  $r^2$ : coeff. of determination,  $N_{\text{Cal}}$ ,  $N_{\text{Val}}$ : number of data pairs in the calibration and

552 validation subsets, respectively, NMSE: normalized mean-square error, FAC2: number of cases when  $p_i/m_i$  is out of the  
553 range of 0.5–2.

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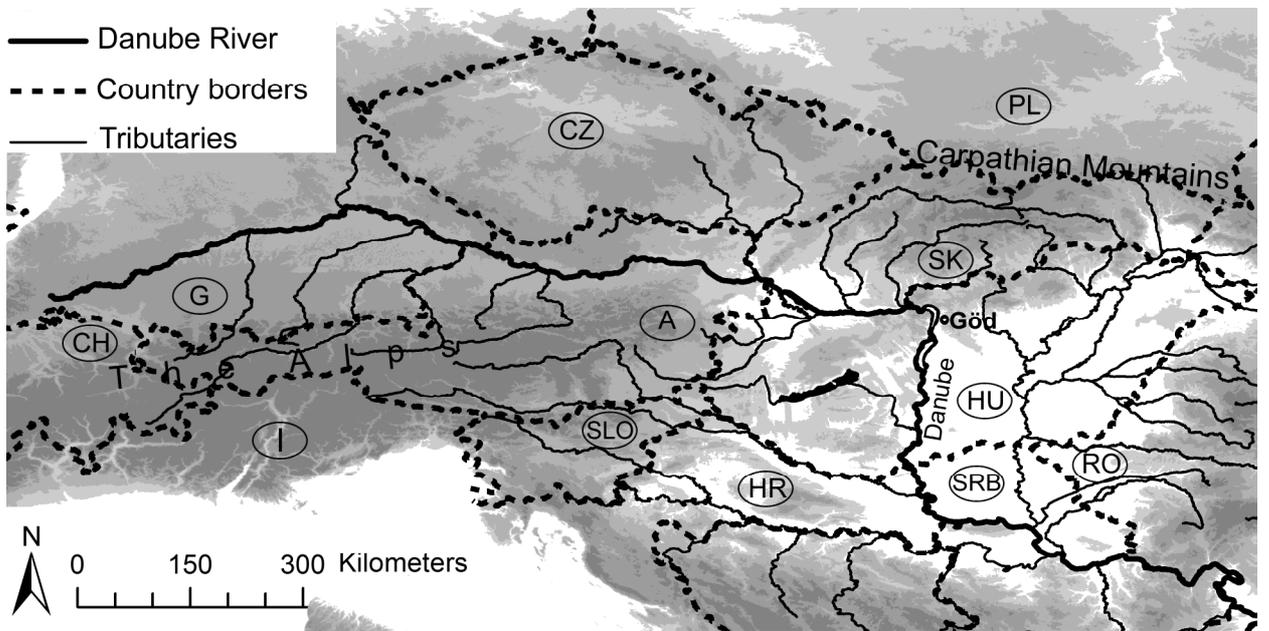
556 Table 5. Regression coefficients of the sediment rating curves for the Danube River at Göd (1668 river km), Hungary,  
 557 between 2003–2011, and model efficiency measures.

Subset	$\log c = a \cdot \log Q + b$		$\log c = a \cdot (\log Q)^2 + b \cdot \log Q + d$			CF	NS	$\overline{ D }$	$\overline{ D }_s$	$r^2$	$N_{\text{Cal}}$	$N_{\text{Val}}$	NMSE	FAC2
	a	b	a	b	d									
Fall	T1		-0.5816	5.0268	-8.8201	-	0.47	54.0	30.0	0.51	24	23	0.64	3
	T2		-0.8659	6.9265	-12.04	-	0.58	48.0	44.6	0.56	41	40	0.23	11
	T3		-0.0168	0.7808	0.9161	1.06	0.25	32.1	32.1	0.29	50	49	0.20	2
Rise	T1		2.0786	-11.387	16.279	-	0.74	21.3	21.3	0.49	19	9	0.04	0
	T2	1.4776	-3.495				0.78	29.7	29.7	0.66	25	25	0.21	0
	T3	1.1168	-2.1849				B-M	0.49	29.7	0.48	27	26	0.16	0

558  
 559 Subsets: T1: 0–5 °C, T2: 5–15 °C, T3: 15–25 °C. c: suspended particulate matter (SPM) concentration ( $\text{mg L}^{-1}$ ). Q:  
 560 discharge ( $\text{m}^3 \text{s}^{-1}$ ). a, b, d: regression coeff. CF: correction factor (B-M refers to Bradu Mundlak, individual value for all  
 561 predicted SPM concentrations), NS: Nash-Sutcliffe efficiency criterion, D: Difference between predicted and observed  
 562 concentration.  $D_s$ : Difference between predicted and observed concentration for those periods when the SPM  
 563 concentrations were  $\geq 5 \text{ mg L}^{-1}$ .  $r^2$ : coeff. of determination,  $N_{\text{Cal}}$ ,  $N_{\text{Val}}$ : number of data pairs in the calibration and

564 validation subsets, respectively, NMSE: normalized mean-square error, FAC2: number of cases when  $p_i/m_i$  is out of the  
565 range of 0.5–2.

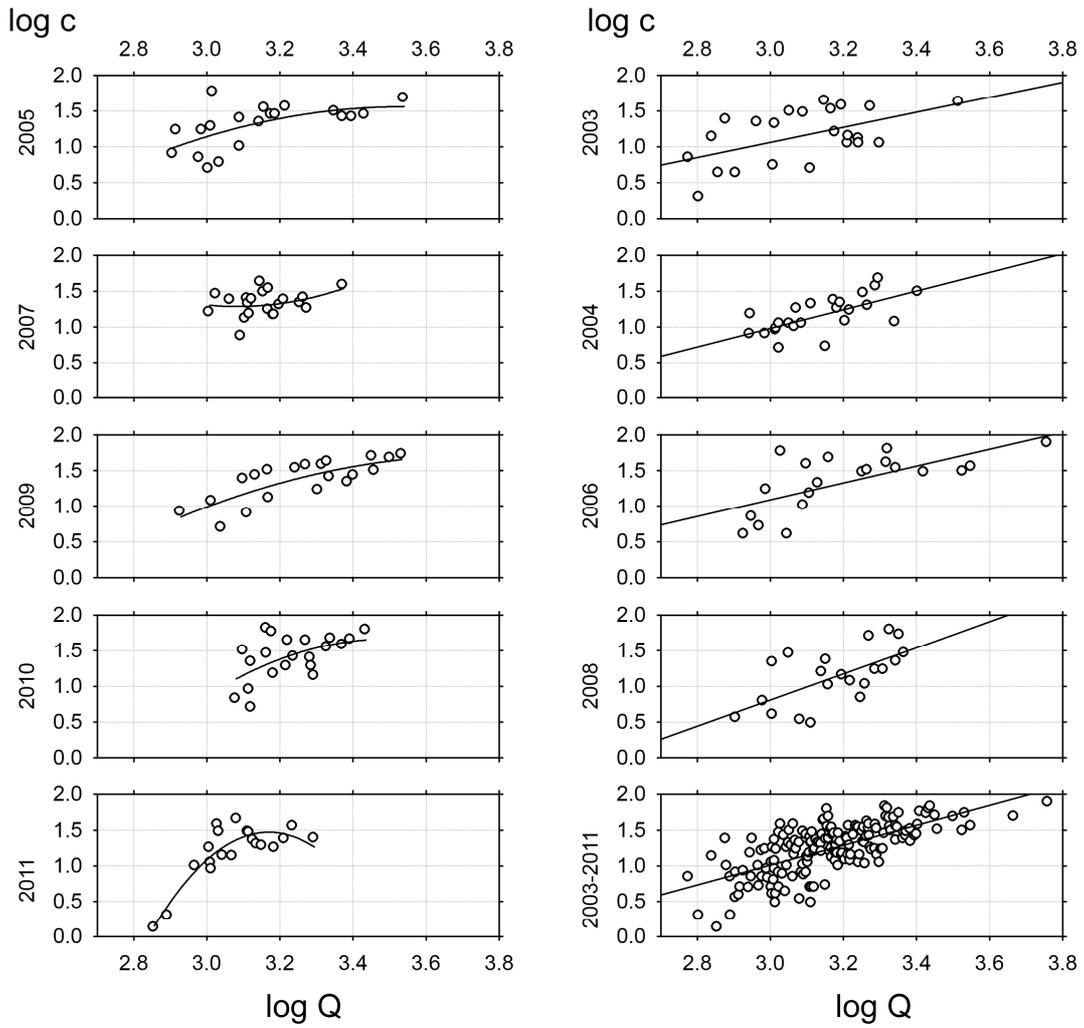
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567 Fig. 1.

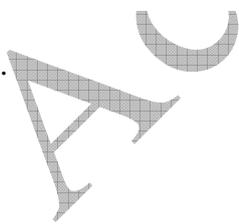
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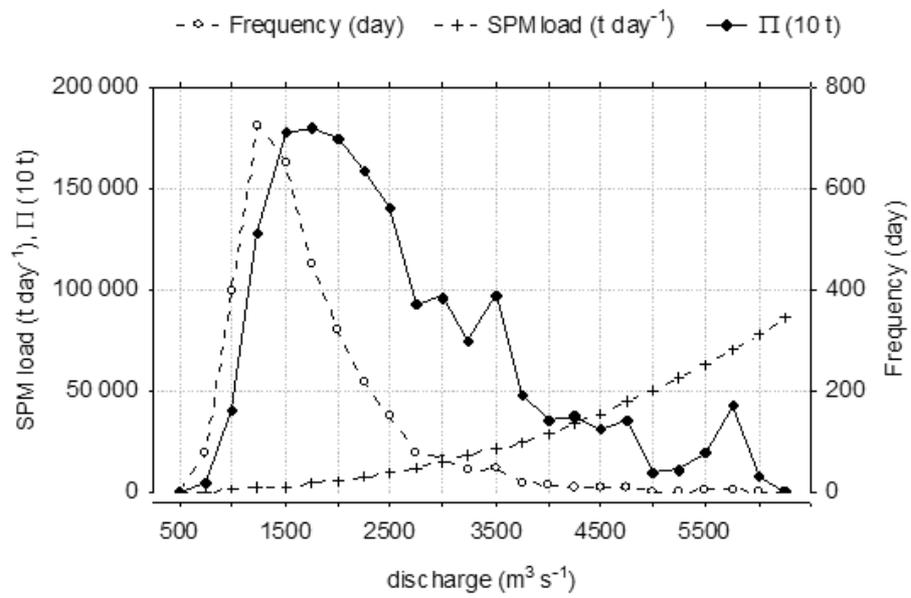


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Fig. 2.

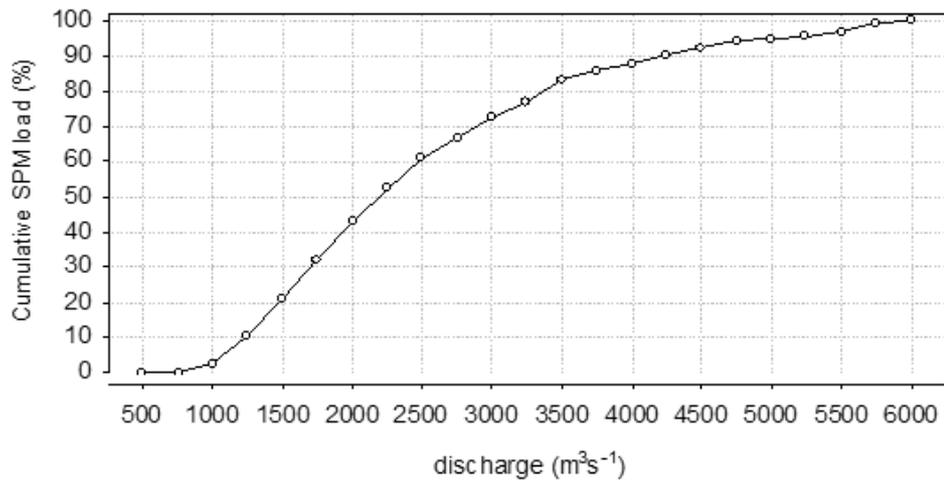




570 Fig. 3.

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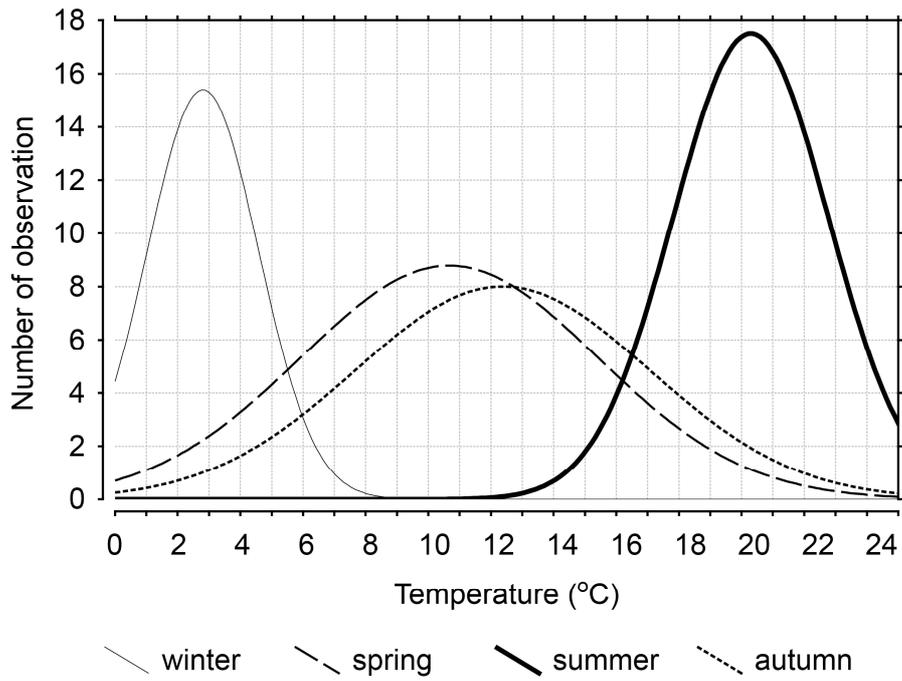


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577 **Figure captions**

578 Figure 1. Map of the Danube catchment area, with the gauging station at Göd (1668  
579 river km).

580 Figure 2. Annual sediment rating curves at Göd, Danube River, Hungary. 'Q' stands for  
581 water discharge ( $\text{m}^3 \text{s}^{-1}$ ), 'c' for the suspended particulate matter concentration ( $\text{mg L}^{-1}$ )  
582 of the water.

583 Figure 3. Flow-frequency distribution, suspended particulate matter (SPM) load, and the  
584 product ( $\Pi$ ) of the instantaneous data at Göd (Danube River, Hungary), 2003–2011. The  
585 highest  $\Pi$  value shows the *effective discharge* of the river.

586 Figure 4. Cumulative suspended particulate matter load expressed as percentage plotted  
587 versus discharge in the Danube River at Göd (1668 river km), Hungary, 2003–2011.

588 Figure 5. Distribution of water temperature data in different seasons in the Danube  
589 River during 2003–2011, at Göd (1668 river km), Hungary.

590