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Selecting a calculation method to estimate sediment and nutrient loads in streams: Application to the Beaurivage River (Québec, Canada)

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Abstract

Estimation of sediment and nutrient loads is of crucial interest for a good assessment of water pollution. This paper proposes an overview of existing estimation methods and a framework to select the most suited one given available streamflow and concentration data. Correlations between contaminant concentration and streamflow should first be checked to generate missing concentration values by regression. However, correlations are not always strong, in which case the ratio estimator method is more appropriate. Given a 6-year data set (1989–1995) from the Beaurivage River (Québec, Canada) with, at best, a weekly sampling, the ratio estimator method was selected to estimate annual and seasonal loads of sediments and nutrients (N and P). Results show relatively steady annual loads (on average 8.1 and 1.1 kg ha yr^{-1} for total dissolved N and total P, respectively) and a low erosion rate (0.23 t ha yr^{-1}). The results also confirm that nutrient and sediment transport via runoff is essentially a springtime process in this region, and they indicate that dissolved P represents the bulk of the total P load, most likely due to artificial subsurface drainage systems in the watershed. These results are compared to the results obtained by using averaging methods and to several other sources of information from literature, confirming the order of magnitude but highlighting some remaining uncertainties. Finally, some research avenues are proposed to improve the proposed framework and to investigate other estimation methods adapted to data characteristics.

Keywords: Water quality; Load estimation; Streamflow; Sediment; Phosphorus; Nitrogen; Beaurivage River

1. Introduction

Water quality is a major concern all around the world, as water uses are threatened by generalised

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contamination resulting from human activities. This contamination concerns sediments as well as chemicals and microbiological components coming from industrial, municipal or agricultural point and non-point sources of pollution. Here non-point pollution refers to sources that are driven by multiple factors and includes diffuse pollution which is exclusively a result of human land use by man and land use changes (Novotny, 1999). In Canada, intensification of agriculture in the last decades has been singled out as the most important non-point source of water pollution. This mainly concerns nutrients nitrogen (N) and phosphorus (P) which are transported from fertilized agricultural lands to surface waters via runoff and erosion and accelerate the eutrophication process. However, this diffuse pollution is also the most difficult to control. This is why disease prevention and environmental protection require intensive monitoring and accurate assessment of water quality in rivers. In environmental management and research, water quality data become imperative to:

(i) assess the water quality status;
(ii) study the controlling processes of water pollution;
(iii) define and apply environmental objectives to restore or improve water quality, such as the Total Maximum Daily Load (*TMDL*) approach (USEPA, 1999; Rousseau et al., 2002) and the combined approach of the European Union Water Directive Framework (Rousseau et al., 2004);
(iv) assess the effects of best management practices (*BMPs*) in a watershed (e.g. Meals and Hopkins, 2002); and
(v) calibrate hydrological and water quality models (e.g. Duan et al., 2003).

While discrete concentration data are useful for comparison to existing water quality standards, they are not sufficient. Indeed, from a management point of view, calculation of mass balances at the watershed scale and assessment of their inter-annual variability require estimation of pollutant loads over long periods (e.g. month, season or year). The information needed for load calculation are streamflows and pollutant concentrations, which are both time- and space-dependent. In most cases, stream gauges continuously monitor streamflow while pollutant concentrations are measured less frequently because of high costs of sampling and laboratory analyses. The challenge is then to make the best use of available data to estimate monthly, seasonal or annual loads.

To meet this goal, several statistical or empirical methods were developed in the early 1980 s and have been used in two ways:

(i) given existing data, finding the most appropriate method to estimate pollutant loads, and
(ii) prior to sampling, defining the optimal sampling strategy and calculation method to minimise number of samples and obtain acceptable load estimates.

This paper reviews existing calculation methods and proposes a practical framework to choose the most accurate method given available data. Then it presents an application of this framework using data from the Beaurivage River watershed (Quebec, Canada) to calculate seasonal and annual loads of sediments and nutrients (N and P). These contaminants were chosen since they are of particular concern in this region. Indeed, high frequency sampling performed in agricultural watersheds of south-western Quebec showed that the total P standard ($0.03\ \mathrm{mg\ L^{-1}}$) was exceeded in most samples and that mean N concentration was increasing (Coote and Gregorich, 2000).

2. Methods for load estimation

The actual load of sediments or pollutants transported through a river cross-section during a time interval is given by:

$$L = \int_{t_1}^{t_2} Q(t)C(t)\mathrm{d}t \tag{1}$$

Where L represents the load between time t_1 and t_2, *Q(t)* the streamflow at time t, and *C(t)* the sediment or chemical concentration at time t. When t is expressed in seconds, Q in $\mathrm{m^3\ s^{-1}}$ and C in $\mathrm{mg\ l^{-1}}$, the result of L is in g. Since streamflows and concentrations are not always measured simultaneously and continuously, the question is to find an estimator of L.

Existing methods for load estimation can be classified in four classes: (i) averaging estimators which are based on a selection of available data, (ii) ratio estimators, (iii) regression methods which use a regression relationship between C and Q (i.e. the so-called rating curve) to estimate unobserved concentration data; and (iv) planning level load estimators.

2.1. Averaging estimators

Averaging estimators, also called integration or interpolation methods, use the means of concentrations and flows over a time interval. Walling and Webb (1981, 1988) defined and analysed six procedures. The four principal procedures are presented below.

The first procedure calculates separately the mean daily flow and the mean daily concentration based on data only from days when both variables are measured:

$$L_s = \frac{\sum_{i=1}^{n} A_i C_i}{\sum_{i=1}^{n} A_i} \frac{\sum_{i=1}^{n} A_i Q_i}{\sum_{i=1}^{n} A_i} n = \bar{C} \cdot \bar{Q} \cdot n \qquad (2)$$

where A_i represents the indicator for availability of concentration data (1 if data is available, 0 if not), C_i the concentration on day i, Q_i the average flow on day i, n the total number of days for the period of load estimation. Overbars denote sample arithmetic means, and L_s the resulting load.

In the second procedure, the mean flow $\bar{Q}$ is replaced by the mean of all flow measurements μ_q. Unlike the first procedure, this one uses all the available data.

$$L_w = \frac{\sum_{i=1}^{n} A_i C_i}{\sum_{i=1}^{n} A_i} \frac{\sum_{i=1}^{n} Q_i}{n} n = \bar{C} \cdot \mu_q \cdot n \qquad (3)$$

with:

$$\mu_q = \frac{\sum_{i=1}^{n} Qi}{n}$$

L_w denotes the load. Walling and Webb (1981), as well as Ferguson (1987), found that these two estimators are precise (i.e. give similar results with different subsamplings from the same data set) but biased (i.e. resulting in a strong underestimation of the actual load). In the third procedure (so-called sample mean), the load is first calculated on each day where both variables are measured, and then the mean daily load, L_a, is given by:

$$L_a = \frac{\sum_{i=1}^{n} A_i C_i Q_i}{\sum_{i=1}^{n} A_i} n = \overline{CQ} \cdot n \qquad (4)$$

This procedure produces a large bias when concentration data are sparse. To reduce the bias, Dolan et al. (1981) replaced C_i by a mean concentration value over a defined period of time.

Finally, the fourth procedure, derived from Eq. (4), weighs the mean daily load by the mean of all measured flows and estimates the ensuing load, referred to as L_c, as follows:

$$L_c = \frac{\dfrac{\sum_{i=1}^{n} A_i C_i Q_i}{\sum_{i=1}^{n} A_i}}{\dfrac{\sum_{i=1}^{n} A_i Q_i}{\sum_{i=1}^{n} A_i}} \frac{\sum_{i=1}^{n} Q_i}{n} n = \overline{CQ} \frac{\mu_q}{\bar{Q}} n \qquad (5)$$

These last two procedures, Eqs. (4) and (5), were found to be less biased than the first two, Eqs. (2) and (3), but resulted in a large variability in load estimations (Walling and Webb, 1981).

These four averaging estimators are widely used as a first approximation. However, if the data set does not represent the entire range of flows and concentration values, bias can be important (Dolan et al., 1981; Ferguson, 1987). Moreover, they are defined for periodic concentration data. For aperiodic data, more suited methods have been developed (see Littlewood et al., 1998).

2.2. Ratio estimators

These methods are derived from the ratio estimator developed by Beale (1962), which multiplies the previous estimator (L_c, Eq. (5)) by a ratio, which accounts for the covariance between load and streamflow values.

$$L_{\mathrm{re}} = \overline{CQ}\,\frac{\mu_{\mathrm{q}}}{\bar{Q}}\,n\left(\frac{1+\frac{1}{n_{\mathrm{d}}}\frac{S_{CQ}}{\overline{CQ}\cdot\bar{Q}}}{1+\frac{1}{n_{\mathrm{d}}}\frac{S_{Q^2}}{\bar{Q}^2}}\right) \tag{6}$$

with

$$n_{\mathrm{d}} = \sum_{i=1}^{n} A_i$$

$$S_{\mathrm{CQ}} = \frac{1}{n_{\mathrm{d}}-1}\left(\sum_{i=1}^{n} A_i C_i Q_i - n_{\mathrm{d}}\bar{Q}\overline{CQ}\right)$$

$$S_{Q^2} = \frac{1}{n_{\mathrm{d}}-1}\left(\sum_{i=1}^{n} A_i Q_i - n_{\mathrm{d}}\bar{Q}^2\right)$$

These unbiased estimators are well suited for cases when a large number of flow data but only a few concentration data are available. Preston et al. (1989) presented several of these estimators, derived from Eq. (6). As reported by Richards and Holloway (1987), the ratio estimator was mandated for use in the Great Lakes region (Canada–US) by the International Joint Commission.

2.3. Regression methods

Regression methods, and their resulting rating curves, define an empirical relationship between streamflow and concentration. The most common regression equation is the log-log linear rating curve:

$$\log_{10}(C) = a + b\cdot\log_{10}(Q) \tag{7}$$

Once fitted to available data by least square regression, Eq. (7) may be used to generate daily concentration values, and then to calculate any resulting load by summing, over a specific period, the product of daily concentration and daily streamflow (Eq. (8) for n days):

$$L_r = \sum_{j=1}^{n} C_j\cdot Q_j \tag{8}$$

The estimator L_r is precise but has been shown to produce a strong underestimation of the actual load when using the log transformation. Ferguson (1987) proposed corrections to get an unbiased estimator L_{cr}:

$$L_{\mathrm{cr}} = L_{\mathrm{r}}\cdot\exp(2.651\cdot s^2) \tag{9}$$

where s indicates the estimated standard error of the estimate of the rating curve in $\log_{10}$ (mg l^{-1}) units.

This method does not require extensive data but the quality of prediction depends on the quality of the correlation between flows and concentrations. This requirement is often met for sediments, particulate and total P, as well as pesticides, but more rarely for mobile chemicals such as nitrate or chlorides (Robertson and Roerish, 1999; Vieux and Moreda, 2003). This relationship closely depends on land use, since important urban areas within a watershed lead to independent dynamics of concentrations and streamflows (Naden and Cooper, 1999). Sometimes, even for agricultural watersheds, no discernable correlation can be found for P because of too sparse data, influence of fertilization events or important dilution effects (Mukhopadhyay and Smith, 2000). Regarding accuracy of load estimation, Walling and Webb (1981, 1988) performed a rigorous evaluation of these regression methods and showed that they can produce an underestimation of 23–83% of the actual load. Dolan et al. (1981); Preston et al. (1989) found average biases of less than 10% for annual loads of total P when using 24 samples a year. It should be noted that the accuracy of prediction also depends on the range of streamflow data used for the regressions. Indeed, the regression equation should be used for interpolation but not for extrapolation. For instance, if the regression equation is based only on base flow values, it cannot be rigorously used to generate peak flow concentration values.

More sophisticated methods have been developed or are currently under development. First, as the temporal variability of the relationship between concentration and streamflow can be very important (Haygarth et al., 2004), some authors proposed to define a regression equation as a function of time in order to take into account nonlinearities as well as seasonal and long-term variability (Cohn et al., 1989). Some authors also proposed to consider the percentage of crop and urban areas on the watershed in the regression equation (Naden and Cooper, 1999), but this requires extensive calibration on several watersheds. It is also possible to use a variable that gives a better correlation with concentrations than streamflow, for example turbidity (Thomas and Lewis, 1995). Finally, Sivakumar and Wallender (2004) developed a non-linear deterministic desegregation approach to estimate sediment loads at high resolution based on low-resolution data. However, this method has not been compared yet to the ‘classical’ methods.

2.4. Planning level load estimation

This method is separate and distinct approach from the others introduced in this section, as it does not need site-specific data either for runoff or concentration. The load is estimated as the product of an estimated cumulative runoff volume and a single representative value of concentration. Due to its simplicity and despite its large uncertainty, it is widely used as a first estimation of pollutant loads, especially for ungauged watersheds (Schwartz and Naiman, 1999). Obviously, if streamflow and concentration data are available, other methods should be used for a better estimation.

2.5. Stratification

Stratification can be applied to the first three methods described in Sections 2.1–2.3. Here, the idea is to classify streamflow values into homogeneous classes and calculate the estimators separately for each class. The choices of the number and size of the classes are crucial and depend on the number of data and the flow characteristics of the river. Many authors use only two classes: low flows and high flows (Dolan et al., 1981; Preston et al., 1989), or sometimes three classes (base flow, high flow, and flood flow) when numerous data are available (Michaud et al., 2004). This approach generally provides a mean to increase accuracy and precision of estimation but can only be applied with a large number of data. Intervals for stratification schemes can also be statistically determined on the basis that distribution of flow data follows a lognormal distribution (Rousseau, 1985; Rousseau et al., 1987).

2.6. How to choose the most accurate method for load estimation?

Few studies have assessed the precision and accuracy of these different methods by comparing the estimators with the ‘true’ or ‘actual’ loads calculated on the basis of a daily or hourly sampling data set. Most of them show that different methods give different results when applied to the same data set. For instance, Webb et al. (1997) showed that, using seven different methods, there could be more than a 50% difference in the magnitude of calculated load. Mukhopadhyay and Smith (2000) found that the stratified sample mean method gave better results for P load estimation than the ratio estimator and the sample mean, even if the results varied widely during their 6-year study period. On the other hand, Dolan et al. (1981) found that the stratified Beale’s ratio estimator gave the best results for the estimation of total P load from the Grand River to Lake Michigan, as compared to five averaging estimators and four regression methods. They recommend this method for total P load when concentration data are limited and a daily flow record is available. In the same way, Rekolainen et al. (1991) showed that Beale’s ratio estimator was more accurate than other averaging methods for total P load estimation for two watersheds in western Finland. However, it has never been evaluated for sediments and N.

The regression method seems to be appropriate for large rivers with low daily variation in concentration, although it is often used for small rivers. Robertson and Roerish (1999) reported that regression methods are especially well suited for small data sets assembled over several years. Ferguson (1987) found that a corrected rating curve gave better results than averaging methods, with a lower variability. Using data from long-term, daily sediment-measuring sites within large ($>10^6$ km^2) to small ($<10^3$ km^2) river basins in USA and Europe, Horowitz (2003) showed that the regression method tends to underpredict high and overpredict low suspended sediment concentrations, with larger errors associated with daily and weekly load estimates than with quarterly or greater load estimates. He further stressed that over periods of 20 or more years, a single rating curve based on all available data generated errors of $<1\%$ and that better results could be achieved using individual annual curves. Meanwhile, Walling and Webb (1988) used rating curves on three rivers in Devon, UK, and found that these methods had not led to accurate estimation of sediment load. Phillips et al. (1999) and Webb et al. (1997, 2000) performed extensive evaluation and comparison of load estimation methods in England for the LOIS (Land-Ocean Interaction Study) program using a 15-min frequency sampling for sediment concentration from several watersheds. The results showed that none of the estimation methods investigated gave reliable estimates of the true sediment load, the most accurate being obtained with rating curves. This led the authors to develop and apply a new and specific approach based on the estimation of errors associated with the different load calculation procedures (Phillips et al., 1999).

Obviously, the accuracy of rating curve methods depends on the correlation between flow and concentration (e.g. see Smith and Croke, 2005). Sivakumar and Wallender (2004) state that, even if a reliable relationship is found, the usefulness of rating curves is limited due to the non-linear propagation of error, as small errors in concentration measurements may lead to large errors in load estimation.

More generally, accuracy of estimation methods depends on many factors such as the frequency of sampling, the length of the estimation period (Littlewood et al., 1998), the size of the watershed (Phillips et al., 1999), the behaviour of the contaminants (Richards and Holloway, 1987) as well as human activities (e.g. land use, tillage practices, fertilization, etc.). Therefore, the choice of the best method is time- and site-dependent (Kronvang and Bruhn, 1996) and the way high-flow events are sampled plays a key role in estimator performance since these events often transport the bulk of the annual load in a short-time period.

In conclusion, as stated by Preston et al. (1989) who compared averaging methods, ratio estimators and rating curves applied to a range of different constituents, no single group of estimators is better than the other in all cases. Nevertheless, based on published information, the above review suggests that: (i) averaging methods are accurate only when concentration measurements are available for the entire flow range; (ii) the ratio estimator is less sensitive to river and pollutant characteristics than regression methods but requires more data to achieve the same level of precision, and is robust and unbiased under systematic sampling, as well as under stratified event sampling; (iii) regression methods can give the best results for sediments and total P if streamflow and concentration data are strongly correlated for a wide range of streamflow values. Therefore, the regression method should be used in priority if the correlation is good enough, but what is 'good enough'? The choice of a criteria and a threshold value remains subjective. To select the regression method, the framework proposed in this paper relies on the coefficient of determination (r^2) and a threshold value of 0.5, which means that more than 50% of the variability of concentration should be explained by streamflow. Fig. 1 gives a diagram of the framework and the different steps that were followed in the application presented in Section 3. Of course, this framework can be adapted by changing the threshold value or integrating other estimation methods currently under development.

It should be noted that softwares such as FLUX (Walker, 1998) and CORAL (Littlewood et al., 1998) have been developed for automatic load calculation based on several methods. For instance, FLUX proposes six different methods: one averaging method, two ratio estimators (including Beale's

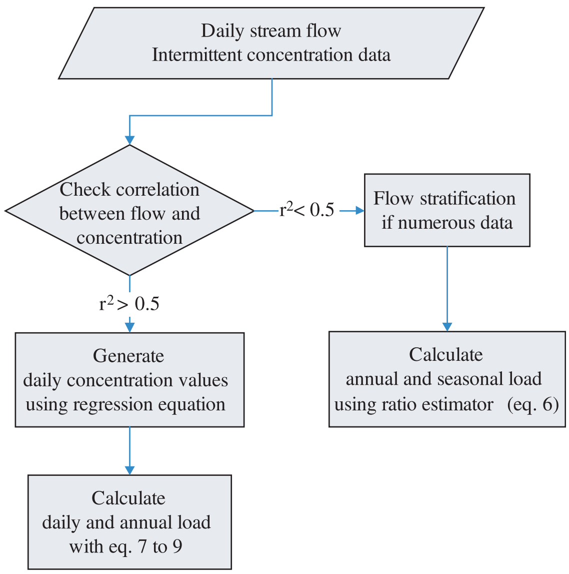

Fig. 1. Diagram of the approach proposed for load estimation.

ratio), and three regression methods. Based on statistical and graphical data analysis, the user selects the most appropriate calculation method and stratification scheme.

3. Application to the Beaurivage River

3.1. The Beaurivage River watershed

The Beaurivage River is a 87-km long tributary of the Chaudière River, located south of Quebec City (see Fig. 2). It drains a watershed of 718 km^2 with 32% used for agriculture, often singled out as an important contributor to non-point sources of pollution. In 1996, Simoneau et al. (1998) reported that the water quality of the Beaurivage River was strongly eutrophicated, the limiting factor being P and 86% of this contribution attributed to agricultural sources. The agricultural activity on the watershed is characterized by significant livestock dominated by pigs (30, 782 pigs, which corresponds to 42.9 pigs km^{-2}, 1994 data, Bédard et al., 1998). Forages (82%), cereals (9%) and corn (8%) represent the dominant croplands.

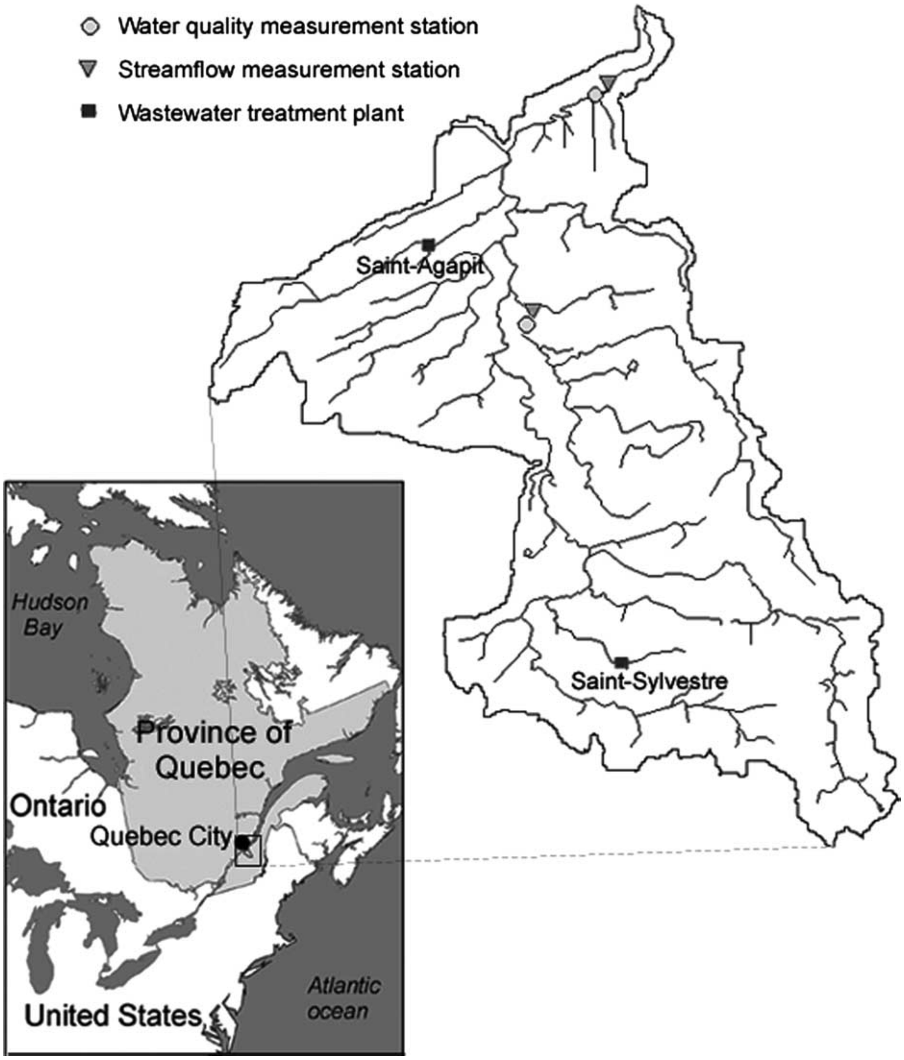

Fig. 2. Location of the Beaurivage River watershed, streamflow and water quality measurement stations, and wastewater treatment plants.

Through the years, repeated applications of large quantities of manure (solid and liquid) on cropland have caused an excessive P enrichment of the soil increasing risks of water contamination (Simard et al., 1995). Bédard et al. (1998) calculated nutrient balances based on organic and mineral fertilization data of 1994 and crop uptake estimation, resulting in exceeding total N and total P of 3,297 t yr^{-1} and 633 t yr^{-1}, respectively. Degradation of water quality is also due, to a lesser extent, to other non-point sources as well as point sources of pollution even though efforts were made over the last decades to control industrial and municipal wastewaters. The population of the watershed is 16,395 inhabitants, but only 7,209 persons are connected to a wastewater treatment plant (1994 data, Bédard et al., 1998). There are two treatment plants within the watershed (see Fig. 2).

To characterize the level and evolution of this contamination, the Quebec Ministry of Environment conducted a water quality sampling campaign between January 1989 and September 1995. Streamflows were monitored daily using an automated gauging station located at Saint-Étienne-de-Lauzon, 5 km upstream of the outlet of the Beaurivage River watershed (see Fig. 3). The area drained at this location is 709 km^2. Water quality was monitored at a station located 1.1 km upstream of the streamflow station, once a week in 1989 and 1990, and twice a month from 1991 to 1995 (see Table 1). For load calculation, sediment and nutrient concentrations were assumed homogeneous in this section of the Beaurivage River. For practical reasons, samples were

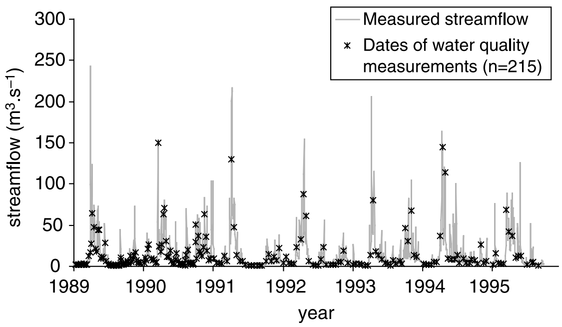

Fig. 3. Measured hydrograph of the Beaurivage River watershed between 1989 and 1995. Asterisks represent the dates of water quality sampling and indicate the corresponding streamflow value.

filtered using 1.2-µm filter instead of the conventional 0.45-µm filter, leading to an overestimation of dissolved forms and an underestimation of particulate forms of chemicals. However, this does not influence the results regarding total P. Water quality parameters were measured following normalised methods: sediment concentration, dissolved P, total P, mineral N (referred to as N–NO_x and including nitrate (NO_3^-) and nitrite (NO_2^-)), ammonium nitrogen (noted N–NH_4), total dissolved N. Particulate N was not measured.

Loads were calculated not only for the whole year but also on a seasonal basis. Fig. 3 introduces the hydrograph as measured at the gauging station, as well as the dates of water quality samplings. As depicted, most peak flow events were sampled for water quality. Unfortunately, no water quality data is available during the highest streamflow measured in spring 1989, and that implies a larger uncertainty in load calculations for that year.

3.2. Estimation of sediment and nutrient load

The first step focused on the assessment of the correlation between concentrations and streamflows to see whether or not the regression method could be used. Table 2 introduces the results of this exercise. The coefficient of determination (r^2) between streamflows and sediment concentrations was 0.43 (data from 1989 to 1995, $n=214$ after excluding one outlier point). This correlation was statistically significant ($p<0.001$, Student t-test). However, this result meant that less than 50% of the variance of sediment concentration could be explained by streamflow which was not good enough to generate concentration data without a large uncertainty. After logarithmic transformation, a r^2 value of 0.14 was obtained from regression between $\log_{10}(Q)$ and $\log_{10}(C)$ (see Fig. 4), corresponding to a r^2 value of 0.25 between Q and C. When fitting a log–log curve by minimizing the root mean square error on C instead of $\log_{10}(C)$, this resulted to a r^2 value of 0.41 between Q and C. This was due to a large dispersion of sediment concentration for base flow. Indeed, when considering only high streamflow values (higher than 9 m^3 s^{-1}), the coefficient of determination between Q and C became 0.65, suggesting that the regression approach could be envisioned for high flows.

Table 1
Number of annual water quality samplings during the study period

	1989	1990	1991	1992	1993	1994	1995	Total
January	4	5	2	2	2	2	2	19
February	4	3	2	2	2	2	2	17
March	4	4	2	2	2	2	2	18
April	5	5	2	2	2	2	2	20
May	4	4	2	2	2	2	2	18
June	4	4	2	2	2	2	2	18
July	5	6	2	2	2	2	2	21
August	4	5	2	2	2	2	2	19
September	5	5	2	2	2	2	1	19
October	6	6	2	2	2	2	1	21
November	4	4	2	2	2	2	1	17
December	3	3	2	1	1	1	1	11
Total	52	54	24	23	23	23	20	218

Table 2
Coefficients of determination (r^2) between all measured variables based on data from 1989 to 1995

	Stream flow	Sediment	DP[a]	PP[b]	Total P	$N–NO_x$	$N–NH_4$	Total dissolved N
Streamflow								
Sediment	0.43							
DP	0.07	0.01						
PP	0.20	0.87	0.20					
Total P	0.18	0.55	0.85	0.65				
$N–NO_x$	0.001	0.01	0.03	0.01	0.02			
$N–NH_4$	0.03	0.01	0.13	0.02	0.09	0.41		
Total dissolved N	0.003	0.01	0.04	0.01	0.04	0.85	0.52	

Note: in some cases, outliers were excluded from calculation.
[a] DP, Dissolved P.
[b] PP, Particulate P.

Regarding P, the bad correlation observed between total P and streamflow ($r^2=0.18$) can be explained by the low variation of P concentration during the year, with a coefficient of variation of 85% (as compared to 160% for streamflow). This is due, in all likelihood, to the non-conservative nature of P, which follows hydrology-independent processes such as biological uptake, and/or to steady-state dissolution kinetic of P. Note that that correlation was not much better for particulate P ($r^2=0.20$). Like for sediments, a large dispersion of P concentration was associated with base flow conditions. The role of point sources of pollution such as wastewater discharge, which is independent from streamflow, could explain this phenomenon. Indeed, the sum of the daily P loads

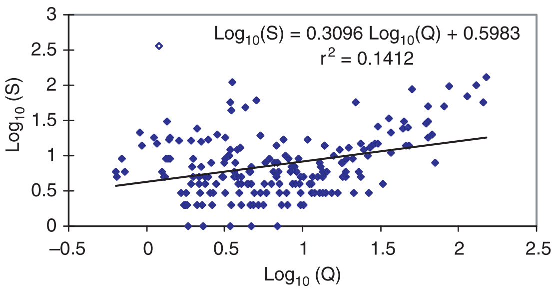

Fig. 4. Sediment concentrations, S, in relation to streamflows, Q, after logarithmic transformation ($n=215$), S: Sediment concentration (mg l^{-1}); Q: Streamflow ($m^3\ s^{-1}$). One outlier (hollow dot) was excluded from correlation calculation ($\log_{10}(Q)=0.08$, $\log_{10}(S)=2.55$).

from the two treatment plants of the watershed was 4.1 kg day^{-1}, which is comparable to the lowest daily load calculated based on measured concentrations and streamflows (3.6 kg day^{-1}). Note that this does not take into account other point sources of pollution (untreated wastewater). When performing the same exercise using one year of data from the Bras d'Henri River, an affluent of the Beaurivage River that drains a small agricultural watershed of 13,306 ha without any point sources of pollution, the correlation between total P and streamflow improved when compared to that for the Beaurivage River ($r^2=0.42$), although it was worse for sediment and streamflow ($r^2=0.29$). Therefore, the presence of point sources of pollution within the Beaurivage River watershed could be a reason for the poor relationship between total P concentration and streamflow. Another factor explaining this result may be the role of artificial subsurface drainage systems (18.1% of cropland area in the watershed, Gangbazo and Babin, 2000), as such systems modify water circulation and induce sediment and nutrient transport during soil saturation conditions. For instance, the study of Yuan and Mitchell (1999) showed that no good correlation could be found between orthophosphate-P and flow from tile flows.

With respect to N, no correlation was found between total dissolved N and streamflow ($r^2=0.004$). This result is not surprising since, unlike P which is a non-mobile chemical mainly transported under particulate form when erosion occurs, mineral forms of N are mobile ions. That means that their transport depends more on their availability in soil solution than on streamflow conditions. Note that particulate N was not measured so that it was not possible to check the correlation between streamflow and total N.

Due to these poor correlations between pollutant concentrations and streamflows, and following the framework of Fig. 1, the Beale's ratio estimator (Eq. (6)) was used to calculate annual and seasonal loads for sediments, N and P. For annual loads, a stratification of data into two classes (low flow/high flow) was performed. The choice of the threshold flow value between the two classes remained subjective and resulted from an analysis of the hydrograph. To get roughly the same number of data in both classes, a threshold value of 8 m^3 s^{-1} was taken, too few data in one class leading to a large uncertainty. For seasonal

Table 3
Annual loads from the Beaurivage watershed as calculated by the ratio estimator method after flow stratification

Year	Water discharge (10^6 m^3 yr^{-1})	Sediment (t ha^{-1} yr^{-1})	DP[a] (kg ha^{-1} yr^{-1})	PP[b] (kg ha^{-1} yr^{-1})	Total P (kg ha^{-1} yr^{-1})	N-NH_4 (kg ha^{-1} yr^{-1})	N-NO_x (kg ha^{-1} yr^{-1})	Total dissolved N (kg ha^{-1} yr^{-1})
1989	367.3	0.09	0.47	0.37	0.85	1.63	4.03	8.28
1990	564.4	0.30	1.16	0.62	1.78	2.37	8.02	12.15
1991	402.0	0.31	0.64	0.55	1.19	1.57	4.43	6.70
1992	363.9	0.30	0.58	0.46	1.04	1.59	4.60	8.17
1993	441.0	0.16	0.35	0.34	0.70	0.83	4.53	6.77
1994	512.7	0.26	0.53	0.47	1.00	1.35	4.01	6.67
Average	441.9	0.23	0.56	0.47	1.09	1.56	4.94	8.12

Note 1: Annual load was not calculated for 1995 since measurements stopped in October 1995. Note 2: These results can be converted to cropland surface by multiplying by 3.04.
[a] DP: Dissolved P.
[b] PP: Particulate P.

loads, no stratification could be done due to scarcity of concentration data (a maximum of 12 measurements per season). This difference in calculation methods implies that annual loads were different from the sum of seasonal loads. Winter loads were calculated from December to February, spring loads from March to May, summer loads from June to August, and fall loads from September to November. Finally, the load calculation for total P was performed based on total P concentration values (sum of dissolved and particulate P). The results were therefore slightly different from the sum of the loads calculated for each form of P.

3.3. Results and discussion

As displayed in Table 3, annual loads of sediments and nutrients were steady during the period of study, except for year 1990 which was characterized by high annual loads of N and P despite relatively low sediment yield. Mean total loads, accounting for non-point as well as point sources of pollution, corresponded to only 29 and 11% of the excess agronomic balances for total N and total P, respectively (see Section 3.1). This difference was most likely due to other sources of pollution (point and non-point) as stated in previous section, but also to the non-conservative nature of N and P suggesting important transformations and losses between cropland and stream network. Seasonal loads of sediments, total N and total P are depicted on Figs. 5–7, respectively. These results confirm that, in this region, water erosion was essentially a springtime process as sediment load calculated for spring season accounted for more than 65% of the annual load every year, reaching 96% during 1991, with a mean of 81% (Fig. 5). This was also true for total P and total dissolved N loads, which followed a similar pattern, even if loads were also substantial in fall seasons of 1990 and 1993 (Figs. 6 and 7). This was most likely due to the large streamflows that were recorded during these two periods (see Fig. 3) even if there was no strong correlation between streamflows and sediments, or nutrient concentrations. Notably, in 1990, the fall load of total P was almost equal to the spring load (0.64 vs. 0.65 kg P ha^{-1}). Moreover, 54% of the mean annual P load could be attributed to the dissolved P load (see Table 3). However, as stated in Section 2, the fact that filtration was performed with 1.2-μm filter instead of the conventional 0.45-μm filter implies an overestimation of dissolved forms of

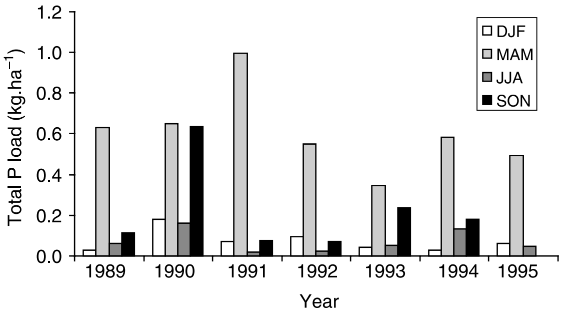

Fig. 6. Seasonal in-stream loads of total P from the Beaurivage River watershed. DJF: December, January, February; MAM: March, April, May; JJA: June, July, August; SON: September, October, November.

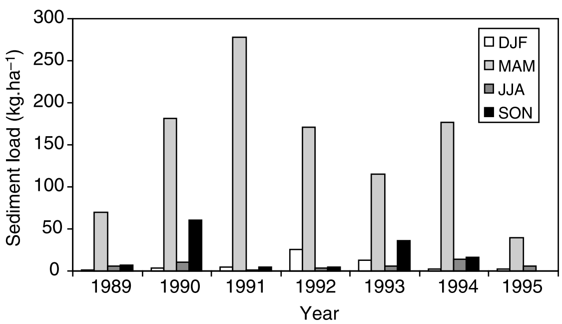

Fig. 5. Seasonal in-stream loads of sediments from the Beaurivage River watershed. DJF: December, January, February; MAM: March, April, May; JJA: June, July, August; SON: September, October, November.

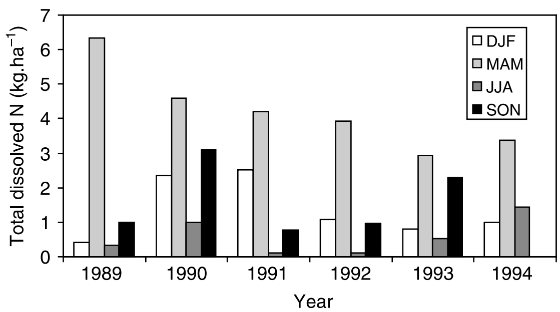

Fig. 7. Seasonal in-stream loads of total dissolved N from the Beaurivage River watershed. DJF: December, January, February; MAM: March, April, May; JJA: June, July, August; SON: September, October, November.

Table 4
Mean relative difference in annual and seasonal values of sediment and nutrient loads calculated for the four averaging methods as compared to the results obtained with the stratified ratio estimator (in %)

Method	Equation	Sediment		Total P		Total dissolved N	
		Annual	Seasonal	Annual	Seasonal	Annual	Seasonal
Non stratified L_{re}	6	−1.9		−0.7		−1.9	
Stratified L_s	2	−50.4	−3.6	−19.2	+2.1	+0.4	+17
Stratified L_w	3	−54.2	−9.9	−25.1	−4.9	−4.9	+4.9
Stratified L_a	4	+2.0	+1.5	+5.9	+3.1	+6.2	+4.9
Stratified L_c	5	−3.0	−3.3	+1.2	−1.9	+4.3	−0.4

chemicals raising awareness in the interpretation of results (but this did not influence total P results). This could also be partly explained by the role of artificial subsurface drainage systems in the watershed as it has been shown to contribute significantly to the transport of dissolved P (Beauchemin et al., 1998; Sims et al., 1998).

Obviously, it is very difficult to assess the accuracy and likelihood of load estimations when there are no 'real' or 'actual' load data available to serve as a reference. However, several sources of information exist that could be compared to the above results: (i) by applying other methods on the same data set; and (ii) by looking at other relevant studies in the literature (i.e. local watersheds that present similar characteristics to the Beaurivage river watershed). Hence, sediment and nutrient loads using the different averaging procedures of Section 2.1, Eqs. (2)–(5), as well as the ratio estimator without flow stratification for annual load, were calculated. Results are given in Table 4 and Fig. 8. The annual loads obtained with the stratified ratio estimator method are generally higher than those obtained with Eqs. (2) and (3), and equivalent or a little smaller than those obtained with Eqs. (4) and (5), both for sediments and nutrients (Fig. 8). Differences are more important for sediment than for total P and are very low for dissolved N. This is probably due to the fact that sediment transport is mainly an event process occurring with peak flow, leading to a strong underestimation of loads when using methods based on the mean measured concentration (Eqs. (2) and (3)). Regarding seasonal loads, the differences are, on average, less important than those for annual loads (see Table 4) but are very variable from year to year (not shown). In some cases, results given by the stratified ratio estimator method are lower than those obtained using Eqs. (2)–(4). This is surprising since Eqs. (2) and (3) have been shown to underestimate actual loads while the ratio estimator is supposed to be unbiased (Dolan et al., 1981). For instance, it is surprising to see that Eq. (2) gives

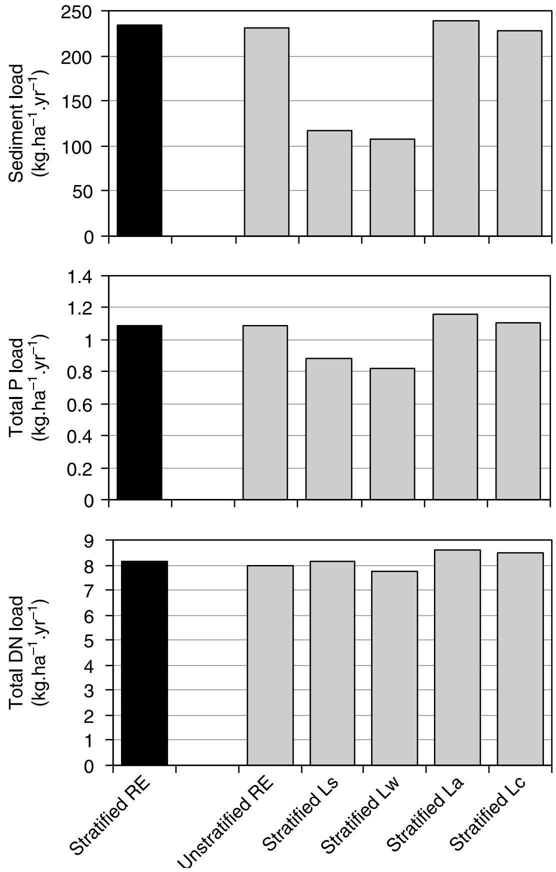

Fig. 8. Comparison of annual loads as calculated with different estimation methods for the Beaurivage river. RE: ratio estimator; DN: Dissolved N.

Table 5
Comparison of estimated mean annual load with those of other relevant studies

Watershed and authors	Estimation method	Sediments (kg ha^{-1} yr^{-1})	Total P (kg ha^{-1} yr^{-1})	Total dissolved N (kg ha^{-1} yr^{-1})	Total N (kg ha^{-1} yr^{-1})
Beaurivage watershed (This study)	Ratio estimator	230	1.1	8.1	
Beaurivage watershed (Bédard et al., 1998)	USLE Eq. (potential soil erosion)	2500			
Beaurivage watershed 1991-1995 (Gangbazo and Babin, 2000)	FLUX ratio estimator	179.3	1.0	7.9	
Nine watersheds in Quebec 1991–1995 (Gangbazo and Babin, 2000) min-max (median)	FLUX ratio estimator	8.6–575.4 (146.5)	0.4–2.2 (1.0)	4.7–23.9 (8.5)	
Binet river watershed 1994–1996 (CART, 1997)	Not specified	191.8	1.5		10
Three watersheds in Lake Champlain basin 1994–2000 (Meals, 2001)	Not specified Flow-based weekly sampling	526–651	1.6–2.7		6.3–8.2 (Total Kjeldahl N)

a mean seasonal load for total dissolved N that is 17% higher than that for the stratified ratio estimator. When looking closely at the data, it appears that this is due to two specific seasonal loads (summer 1993 and spring 1994) for which there were high concentration values for low streamflows, and inversely. Therefore, applying Eq. (2), which multiplies mean concentration by mean streamflow, leads to a very large load value, while applying Eqs. (3)–(5) as well as ratio estimators, which use all streamflow data and daily load means, gives lower results. This is a practical illustration of limitation of Eq. (2) as compared to other equations. The low difference between ratio estimators and other methods may also be attributed to the fact that water quality sampling, even at a low frequency, caught almost all peak flows (see Fig. 3) limiting this way the bias of averaging methods, and making the ratio of Eq. (6) approaching 1. Also, there is very little difference between stratified and non-stratified ratio estimators for annual loads, confirming that stratification is certainly more useful for event sampling data than for regular sampling data. In another study, Gangbazo and Babin (2000) used the FLUX software to calculate annual sediment and nutrient loads with the same data. The selected method was also Beale's ratio estimator, but stratification was performed according to seasons. For the different forms of N and P, their results for mean annual loads are lower but very close to those presented in this paper, with a relative difference always lower than 9%. Surprisingly, for sediments, estimated mean annual loads were 22% lower than those introduced here (see Table 5), emphasizing the importance of the choice of estimation and stratification methods.

With respect to soil erosion in the Beaurivage River watershed, Bédard et al. (1998)used USLE (Wischmeier and Smith, 1978) to estimate the potential soil loss rate. They calculated a value of 2.5 t ha yr^{-1}, which is considered to be a relatively low erosion rate. This is of course drastically larger than the maximum value of net soil erosion calculated in this study using measured sediment concentration data (0.31 t ha^{-1} yr^{-1} in 1991). This corresponds to a sediment delivery ratio (ratio between mean annual net erosion and potential erosion) of 9.2%, which is the expected order of magnitude for large watersheds (10% was suggested as an average value by Roehl

(1962) for watershed of 64,000 acres (i.e. 25,900 ha or 259 km^2).

Another source of information for comparison is data from other watersheds located in the same region. These data are all estimations and are presented in Table 5 with the names and sizes of the watersheds and methods used for estimation. For instance, Gangbazo and Babin (2000) calculated sediment and nutrient loads for nine agricultural watersheds in Quebec using the Beale's ratio estimator method. For sediments, total dissolved N, and total P, the mean annual loads varied widely from one watershed to another, with median values of 0.15, 8.5, and 1.0 t ha^{-1} yr^{-1}, respectively. These values are very similar to those presented here. Other data sets that can be used for comparison relate to the small Binet stream, located in the Chaudière River watershed and draining drains an area of 4.8 km^2 with characteristics similar to the Beaurivage River watershed (CART, 1997), and three subwatersheds of the Lake Champlain basin (Meals, 2001). Even if a rigorous comparison is impossible for sediments and different forms of nutrients since methods of filtration were different and estimation methods are not specified, the estimated loads are similar to those calculated for the Beaurivage River (see Table 5).

Nevertheless, even if these estimates are all of the same order of magnitude, it is not possible to affirm that they are within the order of magnitude of the actual loads. Unfortunately, no study was performed to our knowledge in the Chaudiere–Appalaches region to estimate 'true loads' based on high frequency sampling. Moreover, comparison with true loads from other regions or countries would be irrelevant as erosion and pollutant transport processes closely depend on local characteristics. Therefore, it is difficult to know whether the estimated load values are strongly biased or not, and this has important implications whatever the estimated loads are used for. In our case, the results of this investigation will serve as a first approximation, and in addition to concentration data, for calibration of the water quality component (including modelling of soil erosion, nutrient transport and fate) of the integrated modelling system GIBSI (Gestion Intégrée des Bassins versants à l'aide d'un Système Informatisé; Villeneuve et al., 1998; Rousseau et al., 2000, 2004).

Due to this uncertainty, this application case does not enable to valid or improve the proposed framework for the selection of a calculation method. Presently, this framework still contains subjective elements, such as the threshold value of the coefficient of determination above which regression methods can be used, which makes it adaptable to different cases. An application of the different calculation methods on a complete dataset with daily concentration data would be needed to compare their accuracy and set more precisely their conditions of use. It is also important to note that, since the estimation methods presented are not specific to sediments or nutrients, the proposed approach may also be applied to estimate loads of other contaminants such as pesticides and fecal coliforms.

4. Conclusion

Estimation of pollutant loads is of crucial interest to identify water quality status, understand the processes and identify the sources of pollution. The challenge of load estimation consists in making the best use of available information, which are daily streamflow data and intermittent but generally regular concentration data. The proposed framework helps to select a suited calculation method regarding data characteristics. The first step is to verify if any accurate correlation can be determined between streamflow and concentration in order to generate daily concentration values, since this method gives generally the most valuable results. Unfortunately, like in many cases, this relationship was not good enough for the 6-year data set of the Beaurivage River, probably due to the influence of point sources of pollution, tile drains, and the non-conservative nature of N and P. Therefore, the ratio estimator method was used to calculate annual and seasonal loads, using a flow stratification scheme in the case of annual loads. Even if the results of this study are higher or similar to those obtained using other averaging estimators (which are known to underestimate true loads), and even if they seem to be of the same order of magnitude as those calculated on nearby watersheds, some uncertainties remain about the bias of the estimations due to the scarcity of concentration data. This suggests the need for further water quality monitoring using an adapted sampling strategy and for the development of new estimation methods that would be more adapted to low frequency water quality sampling. For instance, it would be interesting to introduce weights to give more or less importance to the concentration values generated by the regression method as compared to measured values, depending on the regression coefficient. As the correlation between sediments (or any pollutant) and streamflows is generally better for high flows than for low flows, another possibility would be, after flow stratification of the data, to apply a regression method only for a high streamflow class, and an averaging method or the ratio estimator for low flow class. Finally, statistical methods should be investigated to better take into account and quantify the uncertainty linked to estimation. This will be investigated in future works.

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