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Dispersion of pollutants in watercourses intercepted by highway BR-050, in the Triângulo Mineiro region, Minas Gerais, Brazil

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ABSTRACT. Models based on the diffusive-advective equations are valuable tools for forecasting of contamination levels downstream from spill points. These models require field parameters for validation. The longitudinal dispersion coefficient E_L, which translates the transport potential of dilute pollutant is amongst the most important parameters. Based on field techniques using saline and fluorescent tracers, this study determined the E_L in three streams intercepted by highway BR-050, all located in the Paranaíba River Basin. The discrepancy ratios (*Rd*) of some experimental methods and equations reported in the literature were analyzed and compared with the standard routing procedure. Comparing the results of the Moment (statistical), the Chatwin, the peak concentration and the crown concentration methods, the lowest discrepancy ratios were obtained for the latter two methods. Ribeiro, Silva, Soares, and Guedes (2010) also showed that the peak concentration and the crown concentration methods produce the lowest discrepancy ratios. The analysis of contamination scenarios clearly illustrates the direct relationship between pollutant transport capacity and water discharges in natural watercourses.

Keywords: Bom Jardim Stream, Jordão river, saline tracer, fluorescent tracer, longitudinal dispersion coefficient.

Dispersão de poluentes em cursos de água interceptados pela rodovia BR-050, na região do Triângulo Mineiro, Minas Gerais, Brasil

RESUMO. Os modelos baseados nas equações de difusão-advecção constituem valiosas ferramentas para previsão dos níveis de contaminação a jusante do ponto de derramamento. Tais modelos necessitam de parâmetros obtidos em campo para sua validação, dentre os quais se destaca o coeficiente de dispersão longitudinal E_L, que traduz o potencial de transporte de um poluente diluído. Por meio da aplicação de técnicas de campo, com o uso de traçadores salino e fluorescente, neste estudo foi determinado E_L em três córregos situados na bacia hidrográfica do rio Paranaíba e interceptados pela rodovia BR-050. Foram feitas análises da razão de discrepância *Rd* de alguns métodos experimentais e equações da literatura em comparação ao método padrão *routing procedure*. Os resultados dos métodos Momentos (estatístico), Chatwin, pico de concentração e coroa de concentração foram comparados e as menores razões de discrepância *Rd* foram obtidas para os dois últimos. Ribeiro et al. (2010) também mostraram que o método pico de concentração e coroa de concentração apresentam *Rd* menores. As análises dos cenários de contaminação ilustram claramente a relação direta que existe entre a capacidade de transporte dos poluentes com a descarga líquida em cursos de água natural.

Palavras-chave: Ribeirão Bom Jardim, Rio Jordão, traçador salino, traçador fluorescente, coeficiente de dispersão longitudinal.

Introduction

Brazil's southeastern region has a high availability of water resources, which gives its natural watercourses a high capacity for the transport and dilution of conservative or non-conservative contaminants. However, many times the self-cleaning capacity of natural watercourses is misused.

Increasing population density in urban centers and the intensification of agricultural production are increasing the competition over water resources and causing major conflicts. In some cases, the access to water resources are a barrier for economic development of a region. Therefore, it has become increasingly important to complete water resource studies that take into account the water quantity and quality on the watershed scale.

According to Porto and Porto (2008), the management process, based on concept proposed by the National Water Resources Policy, should use modeling technologies to rationalize the various uses of water, and also to understand the necessary management measures to promote a suitable future scenario that meets the needs of the population involved. In this regard, numerous decision support systems for watershed-scale management, which include hydrological and water quality models, have been proposed to solve problems resulting from the pollution of natural watercourses (Argent, Perraud, Rahman, Grayson, & Podger, 2009; Paredes-Arquiola, Álvarez, & Solera, 2010; Paredes-Arquiola, Álvarez, Monerris, & Solera, 2010; Zhang, Huang, Nie, & Li, 2011; Sulis & Sechi, 2013; Welsh et al., 2013; Salla, Pereira, Alamy Filho, Paula, & Pinheiro, 2013).

Most of the water quality models for rivers use the one-dimensional diffusion-advection equation to represent the spatial and temporal distribution of pollutant dispersion (which considers only one direction for the flow) In this analysis longitudinal dispersion is critical to the quality of the modeling (Seo & Baek, 2004). Several researchers have been studying the dispersibility of conservative and non-conservative pollutants in watercourses, including Fischer, List, Koh, Imberger, and Brooks (1979), Devens, Barbosa Jr, & Sivla (2006), Ribeiro et al. (2010), Goldscheider, Meiman, Pronk, and Smart (2008), Silva et al. (2009), Soares, Ribeiro, and Guedes (2010), Azamathulla and Wub (2010) and Wolkersdorfer (2012).

The environmental concerns caused by accidental discharges of dangerous pollutants become even more serious considering that most of these pollutants are considered conservative. This means that it is assumed that they do not react with other pollutants, their concentration is not changed by physical, chemical and biological processes, and their concentration in aquatic environments decreases only through advection and diffusion processes.

The Triângulo Mineiro and the Alto Paranaíba region are connected to the main cities through federal highway BR-050. Road transport plays a major role in the region's economic development and according to the State Secretariat for Environment and Sustainable Development, between 2000 and 2007, this road ranked second only to federal highway BR-381 in terms of accidents with hazardous products in the state of Minas Gerais. The watercourses intercepted by highway BR-050 are essential for the public water supply of municipalities and to meet their consumptive and non-consumptive requirements. This makes it very important to prevent environmental impacts caused by accidents involving dangerous substances and to understand the dispersibility of these pollutants.

In this context, the longitudinal dispersion coefficient, E_L, of four watercourses in the area of influence of highway BR-050 was quantified using:

Five well-known methods involving the use of fluorescent tracer (sodium fluorescein and rhodamine) and saline tracer (sodium chloride, NaCl), which are method of moments (Fischer, List, Koh, Imberger, & Brooks, 1979), routing procedure or propagation method (Fischer et al., 1979; French, 1985), Chatwin's graphical method (Krenkel, 1962), peak concentration method (Rutherford, 1994) and crown concentration method (Devens, Barbosa Jr, & Sivla, 2006);

Four empirical equations widely accepted in the scientific community, developed by Vargas and Riquelme (2001), Kashfipour and Falconer (2002 – citada e não referenciada), Devens et al. (2006) and Ribeiro et al. (2010).

Discrepancy analyses were performed for the results of the empirical equations and experimental methods. The routing procedure method was used for comparison purposes.

Based on the longitudinal dispersion coefficients measured in the field, some additional contamination scenarios were evaluated. These investigations focused on the regions downstream from the spill point, where the first water withdraws are officially registered by the Regional Superintendency of Environmental Regulations – Supram. This study provides an overall understanding of the transport and dispersion of potentially conservative pollutants caused by road accidents on BR-050, thus helping to support the water resources management in this region.

Material and methods

Area of study

The coefficient of longitudinal dispersion, E_L, was quantified in reaches of three watercourses near the city of Uberlandia and in a reach of a watercourse in the city of Araguari. These streams have the potential to transport pollutants to sites where there are water withdraw to urban use and therefore are considered critical to understand the possible consequences of a spill accident on highway BR-050.

The Meio, Retiro and Divisa streams (all tributaries of Bom Jardim stream) were located in PN2 area while the Jordão River is located in the PN1 area as shown in Figura 1. Intersections of the federal highway BR-050 and watercourses are represented by red points. The initial point of the studies reaches are indicated by red arrows.

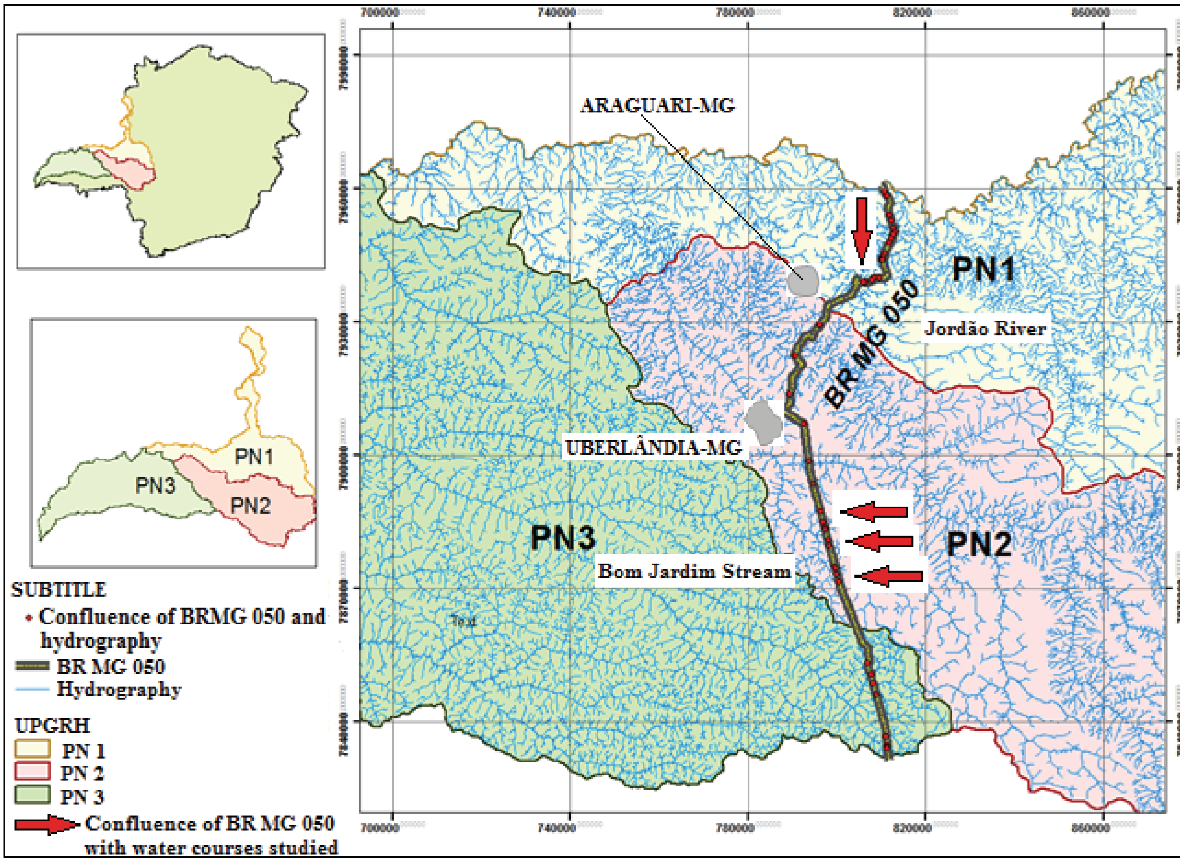

Figure 1. Arrows represent the beginning of the reaches of the watercourses under study and Points represent the intersection of the highway and watercourses.

Methodologies employed to determine E_L

According to Devens et al. (2006), the coefficient of longitudinal dispersion, E_L, is a parameter that indicates the ability of a natural watercourse to disperse a pollutant. However, their determination of the E_L based on empirical equations and experimental methods using a tracer lacks fieldwork and previous laboratory analysis. In fact, geometric and hydraulic data are needed to estimate E_L using empirical equations, such as width of the water surface, average depth of the water, mean flow velocity, bottom slope and velocity shear rate. On the other hand, methods that involve the use of tracers require not only geometric and hydraulic data but also tracer concentration *vs.* time curves in cross sections downstream from the injection point.

This next part of the paper discusses geometric and hydraulic data, tracers, laboratory procedures, the definition of injection and sampling points, sampling procedures, and the experimental methods and empirical equations used in the analysis (or in this study).

Geometric and hydraulic data

Regardless of the methodology employed to quantify E_L, geometric and hydraulic data are fundamental to estimate the dispersion. The geometric parameters were obtained by cross-section bathymetric surveys taken at the injection points at uniform cross sections without backwater or obstacles that could interfere with the longitudinal dispersion of the tracer. The depth at the cross-section were measured with a graduated iron rod, and the widths with a tape measure. The field data was reviewed and entered into excel spreadsheets.

The flow velocities in the cross-sections were measured using a float and a water flow velocity meter. The choice of the most adequate measurement methodology was determined based on watercourse depth. The float was used only in streams less than 0.2 m deep, which limit the use of a water flow velocity meter. The mean flow was calculated using the continuity equation.

The rating curve developed by Salla, Pereira, Alamy Filho, Paula, and Pinheiro (2013) was used for the Jordão River. And the geometric and hydraulic data was estimated based only on the water level measurements.

Tracers used in this study

Soares et al. (2010) states that two basic features characterize a substance as a tracer: (i) it must behave exactly like the medium under study in order to accurately reproduce the dynamics characteristics of the medium, and (ii) it has to be easily detectable in the medium in which it is mixed. Sodium fluorescein (Color Index 45170) and rhodamine (Color Index 45350) fluorescent tracers and sodium chloride (NaCl) saline tracer were used to obtain the dilution and dispersion data in order to monitor and quantify the transit times of the tracer cloud through sampling sections.

Several researchers have studied the applicability of fluorescent tracers in watercourses, including Smith and Pretorius (2002) and Silva et al. (2009). In this study, sodium fluorescein was chosen because it has a low cost and it is easy to detect at low concentrations. Because of its high sensitivity to sunlight, the field work was carried out when there was little sunlight. The samples were stored in dark bottles. Although the use of rhodamine follows the same principle, it is less susceptible to photobleaching. According Moore (2003), Devens et al. (2006) and Goldscheider et al. (2008), as for the saline tracer, although sodium chloride is not an absolutely conservative substance since the type of salt used goes through a dissolution process and part of the ions is retained by the solution. It was chosen due to its low cost, good water solubility, virtual absence in nature and non-toxicity.

Laboratory procedures

The sensitivity of the spectrofluorometer was evaluated before the field work. A serial dilution of the standard solution of 5 ppm of sodium fluorescein tracer was prepared to plot the linear calibration curve and determine the detection limit of the device. The linear calibration curves were obtained by reading the absorbance of the diluted solutions at a wavelength of 492 nm for fluorescein and of 535 nm for rhodamine (10 ppb detection limit). The use of the NaCl saline tracer also required determination of the linear calibration curve to correlate conductivity *vs.* salt concentration. Two portable conductivity meters were used in the field work and the linear calibration curves of the conductivity meters are illustrated in Figure 2. The value of the coefficient of determination (CD) equal to 1 shows good accuracy of the tests.

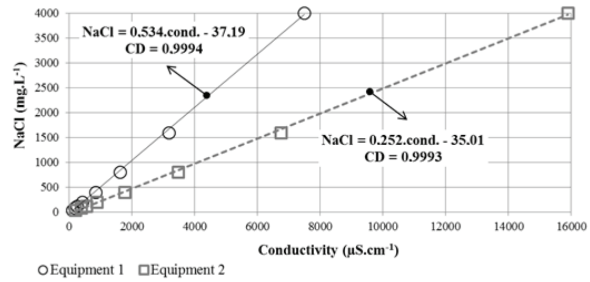

Figure 2. Linear calibration curves of the two portable conductivity meters.

Injection and sampling points

The streams and rivers in the study area are formed mostly of hydromorphic zones, in which surface runoff near the headwaters is intermittent. And this was considered to choose the streams where the studied cold be performed. In order to choose the stream, many sites were previously visited and inspected during the dry period, using as criteria of choice their proximity to federal highway BR-050, the accessibility of injection and sampling points, and the existence of surface runoff during dry season. The first cross-section chosen for sampling was at least at the required distance for complete mixing. In this study, this distance was estimated by Equation 1, proposed by Fischer et al. (1979).

$$L = 0.18*(U.B^2)/(0.6.u*.H) \quad (1)$$

where:
L is the longitudinal length for complete mixing from the injection point (m);
U is the average flow velocity (m s^{-1}),
B is the surface width of the cross section (m),
H is the hydraulic mean depth (m), and
$u*$ is the shear velocity (m s^{-1}).

To determine the amount of tracer injected into the flow that can generate a minimum detectable concentration in the spectrofluorometer and conductivity meter, a simulation was first made using the Gaussian analytic solution of the diffusion-advection equation. Arbitrary E_L values were used to predict the amount of mass to be injected, which promoted a peak concentration exceeding the detection limits of the spectrofluorometer and conductivity meter. Based on the previous simulations of concentration *vs.* time curves for different values of E_L, various scenarios were generated for plume arrival time, rise and fall time, as well as the maximum concentration of tracer in each sampling segment. This allowed for the definition of the sampling distribution and frequency, the number of people to be deployed in each cross-section, and other logistical aspects.

Collection of samples

The tracers were injected instantaneously at the injection point (P_{inj}) by pouring the entire volume into the center of the cross section of the watercourse. After the tracers travelled the entire longitudinal mixing length, water samples were collected at sampling point 1 (P_1) and then at sampling point 2 (P_2). To determine the concentration profile using saline tracer, the probes of the conductivity meters were introduced directly into sampling points P_1 and P_2. To determine the concentrations of the fluorescent tracers, forty samples were collected at the same sampling points and stored in 30-ml dark polyethylene bottles, which were then stored in a dark storage bag. The dark bottles and dark storage bag prevented photobleaching of the samples. This number of collected samples enabled the response curves (or concentration profile, or tracer concentration *vs.* time curves) to be developed.

Empirical equations

In a natural watercourse, the longitudinal dispersion coefficient E_L depends on the width, depth, roughness, flow, and particularly on the flow regime, i.e., turbulence. Ribeiro et al. (2010) found that empirical equations are very useful for predicting E_L, because they make it easy to determine E_L based on flow-related parameters. According to Devens et al. (2006), the use of practical prediction equations is dictated by the logistics required for field work and the need for qualified technical staff. In this study, E_L was estimated by all four empirical equations, which are described in Table 1.

Table 1. Equations used in this study to estimate E_L.

Authors (year)	Equations for estimating E_L
Vargas and Riquelme (2001)	$E_L = 7.3867.(B\ Rh^{-1})^{-1,8558}.(U^2.B^2)/(u\star.H)$
Kashfipour and Falconer (2002)	$E_L = 10.612.(H.U^2)/u\star$
Devens et al. (2006)	$E_L = 0.729.U^{0,774}.B^{1,031}.S^{0,036}.H^{-0,151}$
Ribeiro et al. (2010)	$E_L = 7.326.u\star^{0,303}.H^{1,316}.B^{0,445}.U^{1,458}$

Where: E_L is the longitudinal dispersion coefficient ($m^2\ s^{-1}$); B is the surface width of the cross section (m); H is the hydraulic mean depth (m); Rh is the hydraulic radius (m); U is the average flow velocity ($m\ s^{-1}$); S is the bottom slope ($m\ m^{-1}$); and $u\star$ is the shear rate ($m\ s^{-1}$).

Tracer-based methods

The most suitable way to determine E_L is using tracer-based methods because they correctly simulate the movement and dispersal of the solute, implicitly taking into account all the geometric and hydrodynamic characteristics of the flow. This study evaluated only its longitudinal dispersion (one-dimensional transport).

In these methods, the conventional way to present the response of a flow to the injection of a tracer is to graphically represent the variation in tracer concentration over time (the tracer response curve) detected in two or more cross sections downstream of the injection point, called sampling sections (the distances between the sections in this study are given in Table 1). The tracer response curve, which is defined based on an analysis of water samples collected at selected time intervals during the passage of the tracer cloud, serves as the basis for determining the time course and dispersion characteristics of a pollutant in watercourses. Researcher who have studied the subject include Soares et al. (2010).

The tracer-based methods used in this work were: the statistical method to determine parameter, known as method of moments (Fischer et al., 1979); the routing procedure or propagation method (Fischer et al., 1979; French, 1985); Chatwin's graphical method (Krenkel, 1962), the peak concentration method (Rutherford, 1994), and the crown concentration method (Devens et al., 2006). These methods are widely accepted in the scientific community.

Discrepancy ratio

The comparative method of the discrepancy ratio (*Rd*), originally described by White, Milli, and Crabbe (1973), is used to test the consistency of the experimental data for the longitudinal dispersion coefficient E_L. It is defined as the logarithm of the ratio between the estimated (E_i) and the measured or observed (O_i) values, $Rd = log\ (E_i\, O_i^{-1})$.

The predicted E_L is considered to be identical to the measured value when the *Rd* value is zero. If it is higher than zero, the predicted E_L is overestimated, and if it is lower than zero, the E_L is underestimated. Based on this comparative method, the values obtained by the Routing Procedure were adopted as observed values (O_i), while those obtained by the empirical formulas and by the other four experimental methods were adopted as estimated values (E_i).

The Routing Procedure was chosen as reference because it uses the principle of superposition; hence, it does not take into account how the tracer is released, and is able to adjust the E_L value that compares the field measurements against the concentrations presented by the model. In other methods, such as the method of moments, there area problems related to long tails in the concentration distribution. The Routing Procedure does not present these issues.

Contamination scenarios

The purpose of the scenarios was to verify the contamination of water supplied to the first user downstream from the tracer injection point, based on the accidental spill of a contaminant in the Meio, Retiro and Divisa streams. Simulations of the *C vs. t* tracer clearance curves were completed adopting the E_L values obtained in this study, using experimental methods. And consequently it was possible to determine the concentration of the tracer at critical sections (from where water would be withdraw to the first user).

These studies were performed for the most critical situation, i.e., when the discharge was low. The $Q_{7,10}$ flow (minimum seven-day flow and ten-year recurrence) was considered at the tracer injection point in each watercourse. The $Q_{7,10}$ was determined using contour maps of specific yield (in L s^{-1} km^{-2}), which combines the yield with the drainage area, type of soil, and physical and meteorological characteristics of the watershed. The information about the location of the first user, his distance downstream from the injection point, and the contour maps of specific yield were supplied by the Minas Gerais Institute of Water Management – Igam/Geara and by the Regional Superintendency of Environmental Regulations of Triângulo Mineiro and Alto Parnaíba – Supram/TMAP.

Results and discussion

Table 2 presents the geometric and hydraulic data used for the determination of the longitudinal dispersion coefficient E_L, using equations and experimental methods. While the water discharge into the Jordão River was 7.510 m^3 s^{-1}, the streams showed a much lower capacity for pollutant transport and dilution, i.e., a maximum of 0.072 m^3 s^{-1} in the Divisa Stream, 0.170 m^3 s^{-1} in the Meio Stream, and 0,447 m^3 s^{-1} in the Retiro Stream. Table 2 also shows that the complete mixing length was considered in all the watercourses, starting from the injection point, P_{inj}.

This paper presents only the results obtained with the routing procedure, since this was the standard method of comparison in the discrepancy analysis, as illustrated in Figure 3. The results obtained by the other methods and by the equations described in Table 1 are given in the discrepancy analysis in Figure 4, keeping in mind that an *Rd* of zero indicates similarity with the standard method of comparison.

The same amount of tracer was released instantaneously into all the watercourses, i.e., 5 kg of sodium chloride, 50 g of fluorescein, and 900 g of rhodamine. The highest peak concentrations of tracers were found in the watercourses with the lowest water discharge capacities, indicating their lower pollutant dilution capacity (see Figure 3).

Published works on watercourses with similar geometric and hydraulic characteristics have reported similar results to the streams of this study. In a study in the region of Ouro Preto (Minas Gerais State), Devens et al. (2006) used saline solution as a tracer and reported E_L of 0.143 to 1.290 m^2 s^{-1} for a flow ranging from 0.0052 to 0.1763 m^3 s^{-1}

Analysis of the discrepancy ratio

Figure 4 illustrates the discrepancy ratio *Rd* between the experimental methods and equations from the literature and the standard Routing Procedure. The analysis of the results for each watercourse is not conclusive because the discrepancy differs according to the various methods and equations described in the literature. In general, saline, fluorescein and rhodamine tend to be underestimated by the experimental methods and overestimated by the equations. The highest numerical contrast was found for the Retiro Stream, where the discrepancies of the equations described in the literature (0.302 to 2.569) were higher than those of the various methods, which varied from -0.593 to 0.708 for saline and 0.085 to 0.881 for fluorescein. According to the equations, E_i O_i^{-1} remained below 0.5 and above 2.0, indicating a high discrepancy between the aforementioned equations and the Routing Procedure.

Table 2. Geometric and hydraulic data used for the prediction of E_L.

Watercourse	Tracer	Q (m^3 s^{-1})	A (m^2)	B (m)	H (m)	U (m s^{-1})	P (m)	S (m m^{-1})	$u^\star$ (m s^{-1})	Rh (m)	L (m)	P_{inj} -P_1 (m)	P_1 - P_2 (m)
Meio Stream	NaCl	0.115	0.35	1.85	0.19	0.33	1.97	0.006	0.102	0.175	9.9	87	165
	NaCl Fluorescein	0.170	0.41	1.90	0.24	0.41	2.05		0.109	0.200	9.5		
Retiro Stream	NaCl	0.447	0.49	2.85	0.17	0.91	3.08	0.005	0.091	0.160	78.3	105	192
	NaCl Fluorescein	0.433	0.46	2.80	0.16	0.93	3.03		0.087	0.145	81.8		
Divisa Stream	NaCl	0.072	0.20	0.90	0.22	0.37	1.16	0.004	0.079	0.164	2.9	110	260
	NaCl Fluorescein	0.062	0.16	1.00	0.16	0.39	1.20		0.072	0.136	5.8		
Jordão River	Rhodamine	7.510	12.85	23.04	0.56	0.58	20.44	0.009	0.246	0.628	376.7	495	317

Q = average flow; A = cross-sectional area; B = is the surface width of the cross section; H = hydraulic mean depth (m); U = average velocity; P = wetted perimeter; S = bottom slope; u★ = shear rate; Rh= hydraulic radius; L = complete mixing length from the injection point (equation 1); Pinj = tracer injection point; P1 = first sampling section; P2 = second sampling section.

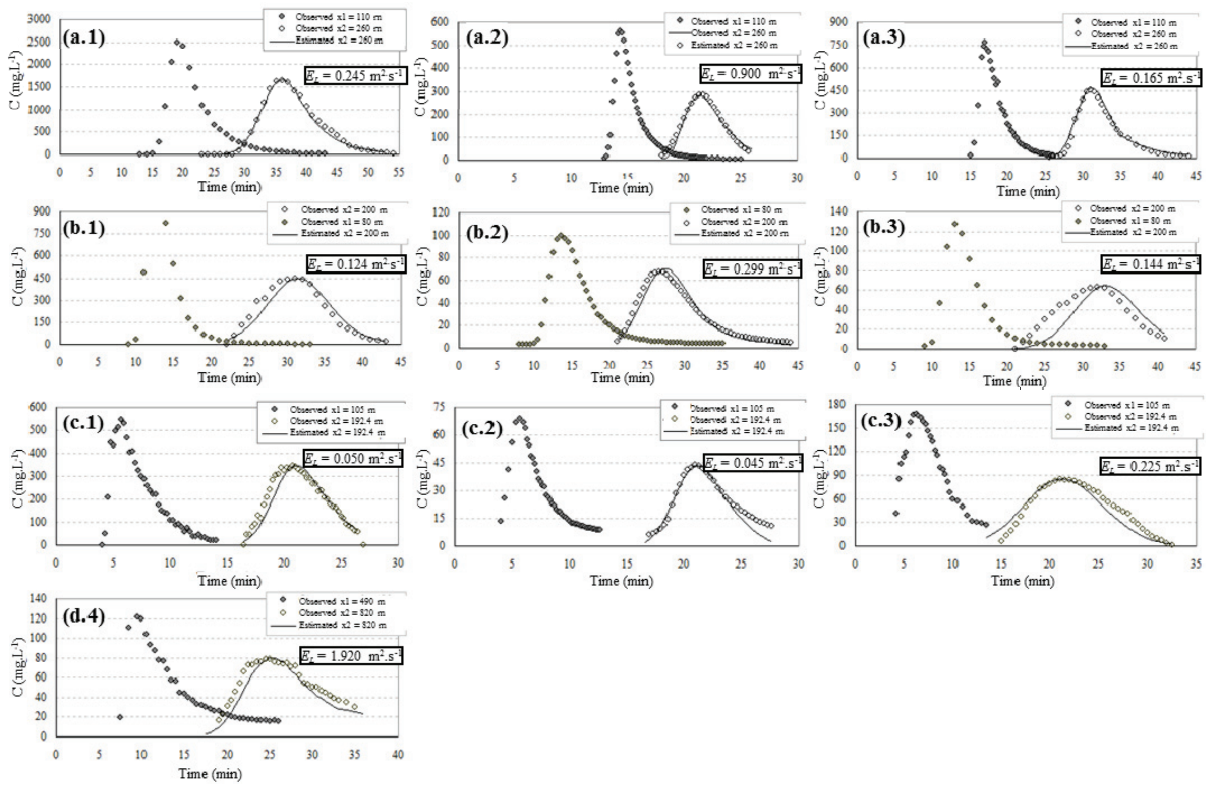

Figure 3. Results of the Routing Procedure for: 1: fluorescein, 2 and 3: saline solution, 4: rhodamine, a: Divisa Stream, b: Meio Stream, c: Retiro Stream, d: Jordão River.

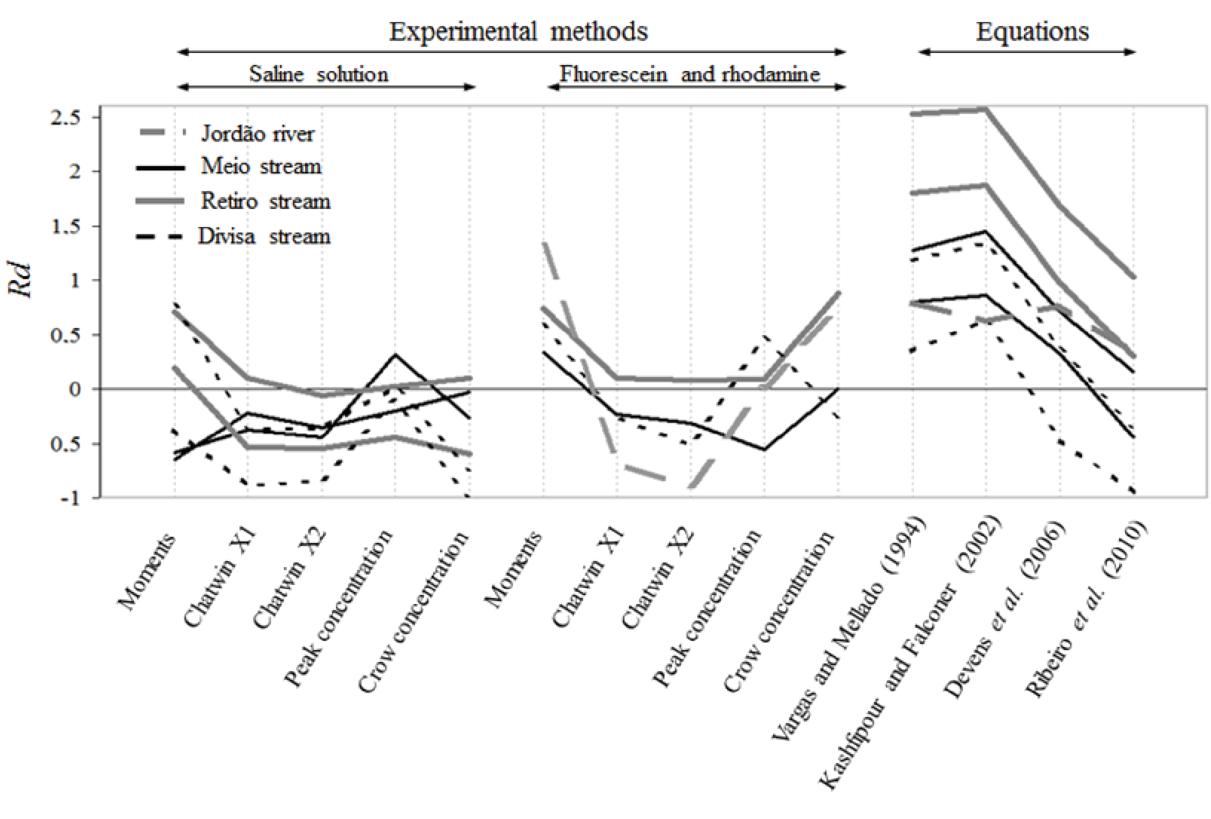

Figure 4. Discrepancy ratio, *Rd*, of the experimental methods and equations reported in the literature *vs*. the standard *Routing Procedure*.

An analysis of the experimental methods revealed that the peak concentration and crown concentration methods yielded the lowest *Rd* values. The crown concentration method yielded an *Rd* value of -0.0035 for in the Meio Stream, while the peak concentration method yielded 0.0131 for the Jordão River and 0.0170 in the Retiro Stream.

Contamination scenarios

According to information obtained from IGAM, the first user downstream from the tracer injection point is located 864 m downstream in the Meio Stream, 1023 m downstream in the Retiro Stream, and 500 m downstream in the Divisa Stream. The peak concentration of tracer in the sections where water is withdraw to the first users is indicated in the tracer concentration *vs*. transit time curves illustrated in Figure 5.

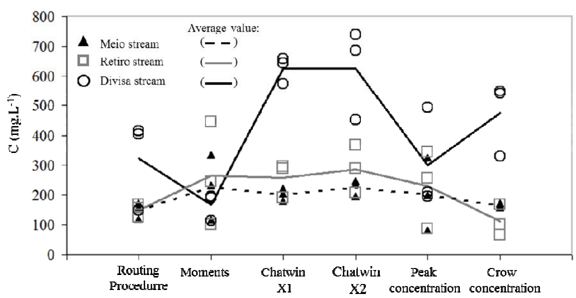

Figure 5. Peak concentration reaching the first user downstream from the P_{inj} in the watercourses under study.

According to most of the experimental methods analyzed in this paper, the low pollutant transport and dilution capacity of the Divisa Stream resulted in the highest peak contaminant concentration, which varied from 113.7 to 740.6 mg L^{-1}. The peak concentrations in the Meio and Retiro streams, according to the different experimental methods, present the same trend in the average curves, indicating higher transport and dilution capacity than the Divisa Stream.

Conclusion

The NaCl injection methodology produced satisfactory results in the experimental determination of the longitudinal dispersion coefficient E_L. This suggests that the methodology can be used as a good replacement for the fluorescent tracer, reducing the cost and time spent on laboratory analyses of fluorescein and rhodamine.

The analysis of the discrepancy ratios, *Rd*, reveals that the experimental methods tend to underestimate saline solution and fluorescein while the equations tend to overestimate them. The peak concentration and crown concentration methods and the equation proposed by Ribeiro et al. (2010) showed the lowest *Rd*.

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