Pollutant dispersion modelling for Portuguese river water uses protection linked to tracer dye experimental data

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Abstract: - Mathematical models are well known as useful tools for water management practices, directly or indirectly related to the implementation of the Water Framework Directive (WFD) in European countries. They can be applied to solve or understand either simple water quality problems or complex water management problems of trans-boundary rivers or multiple-purpose and stratified reservoirs. Accidental spills of pollutants are of general concern and could be harmful to water users along the river, becoming crucial to get knowledge of the dispersive behaviour of such pollutants. In this context, the mathematical modelling of dispersion phenomena can play an important role. Additionally, a judicious selection of mathematical models for application in a specific river basin management plan can mitigate prediction uncertainty. Therefore, intervention measures and times will be established with better reliability and alarm systems could efficiently protect the aquatic ecosystems, the water uses and the public health.

The main purpose of this paper is to evaluate the performance of river water systems dispersion modelling, based on tracer experiments data for calibration and validation. The present work describes the methodology used in the monitoring programs, basically consisting in the injection of a tracer dye (*rhodamine WT*) in an upstream river section and follow-up of the dye cloud along the river to determine the water dispersion behaviour *in situ*. The models were developed to simulate different water quality management scenarios on each reach of the three Portuguese rivers under study: Mondego, Douro and Tagus rivers. However, further developments are needed for Douro and Tagus rivers in order to simulate vertical and transversal dispersion processes and improve the model correlation with the experimental data. The models were calibrated and validated in order to produce operational tools used to estimate the probabilistic leading edge/peak/tail times, the pollutant losses by volatilization, adsorption, precipitation, etc. and remaining concentrations. These tools allows to define, for example, how long water intake need to be suspended after a pollutant spill and can be easily integrated in a future DSS, which should be developed and implemented by each one of the river basin management authorities. The good correlation between experimental and simulated data allows us to conclude that the applied models are accurate enough to describe and predict conservative pollutant transport under different hydrodynamic scenarios. This methodology is appropriate to assess the environmental impact of pollutant loads directly introduced into the streams and, subsequently, to define and implement the best water sources protection practices.

Key-Words: - Dispersion modelling; Water uses protection; Rhodamine; parameter estimation; Portuguese rivers.

1 Introduction

Surface waters, as an important component of the natural environment, need to be protected from all pollutant sources because man's own survival depends on their efficient use.

Related to anthropogenic activities, point and diffuse sources of pollution have driven two important water quality problems in surface waters: eutrophication (excessive nutrient enrichment causing algal blooms) and contamination by hazardous chemical compounds [1].

River hydrodynamics and pollutant discharge dispersion characteristics are determinant factors in river basin planning and management, where different waters uses and aquatic ecosystems protection must be considered [2], namely during the Strategic Environmental Assessment of river basin planning processes in order to preserve the cultural, natural and ecological structures and to promote a sustainable development [3]. The European Water Framework Directive (WFD) [4] establishes a scheduled strategy to reach a good ecological status and chemical quality for all European water bodies.

Mathematical models can be applied in various water management issues, directly or indirectly related to the implementation of the WFD.

According to the *key activity 2* of a strategic EU document [5], models application is specifically indicated for analysis of pressures and impacts (*Project 2.1*) and for water resources monitoring (*Project 2.7*).

The benefits of the synergy between modelling and monitoring are often mentioned by several authors and the linkage of both approaches makes possible to apply cost-benefit measures [6]. Therefore it is essential to correlate monitoring and modelling information with a continuous feedback, in order to optimize both processes, the monitoring network and the simulation scenarios formulation.

Ever increasing computational capacities provide the development of powerful and user-friendly mathematical models for the simulation and forecast of quality changes in receiving waters after land runoff, mining and wastewater discharges [7].

In the context of environmental issues, the probability of occurrence of an adverse effect on human health or aquatic ecosystems due to a hazard exposure is defined as environmental risk [8].

The World Health Organization (WHO) concludes, in the *Guidelines for Dinking Water Quality* (3rd edition), that an integrated management of risks in source waters, treatment systems and distribution networks is the most effective way to guarantee safe drinking water consumers.

An integrated approach (hydrodynamics and water quality issues) is fundamental to prioritise risk reduction options in order to protect water sources and to get a high quality of the raw material for the water supply systems [9]

The main purposes of this research work were parameter estimation for *in situ* characterization of dispersive behaviour in river water and performance evaluation of numerical models, when applied to pollutant transport modelling in three different river systems.

Several tracer dye experiments were carried out in order to improve the knowledge about different water bodies' dispersion patterns and to obtain field data for water quality model calibration and validating procedures.

This approach can constitute a power and useful operational tool to establish better early warning systems and to improve management practices for efficiently protect river water sources and, consequently, public health.

2 Methods

2.1 Tracer dye experiments

Ideally, monitoring networks should be designed to provide a coherent and comprehensive overview of ecological and chemical status within each water body, but this requires an enormous amount of information, monitoring stations and high operational costs.

Integrated models can be used to calculate hydrodynamic parameters and water quality characteristics at any section using data from a monitoring programme applied to limited number of sampling or measuring stations, providing the optimization of the designed monitoring network (Figure 1, [10]).

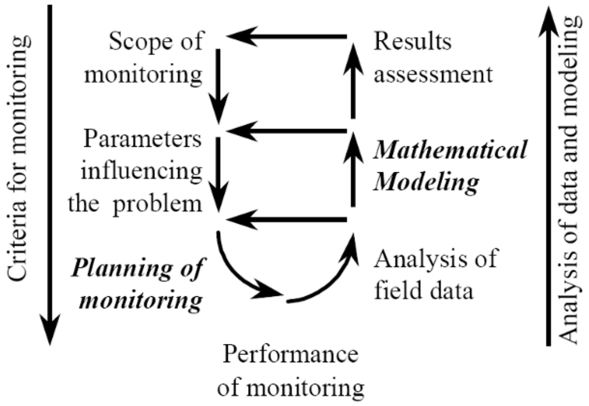

Fig.1: Interaction between monitoring and modelling for monitoring network optimization

The three monitoring programs, each one comprising several sampling campaigns, were carried out using *Intracide rhodamine WT* (Crompton & Knowles Corporation) as tracer dye, recommended for its characteristics: it is non-toxic, non-reactive, has high diffusivity, is highly detectable, and has low sorption and low acidity. Additional characteristics are presented in Table 1.

Table 1: Technical data of *Intracid Rhodamine WT*

Specific gravity	1.15 (20ºC)
pH	10.8±0.7 (20ºC)
Optimum excitation wavelength	≈ 556 nm
Optimum analysing wavelength	≈ 580 nm
Freezing point	≈ -10ºC
Viscosity	< 25 cp (25ºC)

A *Turner Designs* fluorometre was used for tracer concentration measurements. Blanks were taken in all sampling sites for background natural fluorescence determination.

For each case study, the location of the sampling sites was established according to the aims of the respective monitoring program, the accessibility to sampling sites (existence of bridges, availability of boats), river physical characteristics, mixing conditions, weirs location and availability of human resources. Experimental longitudinal dispersion coefficients were calculated from concentration-time curves at consecutive sampling sites, using the methodology described by Chapra [11] for tracer studies.

The amount of tracer dye to inject upstream was calculated by considering the water volume estimated for the river reach or reservoir system and the fluorometre detection limit, because tracer concentrations at the downstream sampling stations must be detectable.

The calculation of the tracer mass recovered at each site allowed the assessment of the importance of physical and biogeochemical transformations in the river stretch under study by quantifying precipitation, sorption, retention and assimilation losses.

The river flow values for each water body and sampling program was selected taking into account different hydrodynamic scenarios: hydropower plant discharge patterns; dry-weather conditions, flood and frequent flows obtained from flow gauge station records [12].

2.2 Mathematical modelling

Mathematical modelling is a powerful and a multifaceted tool to achieve a better understanding of the physical, chemical and biological processes in water bodies, based on a “simplified version of the real” described by a set of equations, which are usually solved numerically.

The formulation of a model requires the better (possible) definition of the geometry and bathymetry of the water body and the interactions with the surrounding processes occurring in its boundaries (boundary conditions).

The models to be used for the implementation of the WFD should ideally have the highest possible degree of integration to comply with the integrated river basin approach, coupling hydrological, hydrodynamic, water quality and ecological modules as a function of the specific environmental issues to analyse.

The *DUFLOW package* [13] was designed to cover a large range of applications in different water systems and to assess water quality problems.

The *DUFLOW* hydrodynamic module is based on the one-dimensional partial differential equations used to describe non-stationary re in open channels. These equations are the mathematical translation of the laws of conservation of mass and momentum and compute flow discharges and elevations at the same point.

The *DUFLOW* water quality module is based on the partial differential equation describing the concentration fate of a constituent in a one-dimensional system as a function of time and place, where a *production* term includes all physical, chemical and biological processes to which a specific constituent is subject to. The process descriptions can be supplied by the user, who can assume different types of kinetics for a specific river model.

The *ADZ* modelling technique (*ADZTOOL*) is a common approach for modelling dispersion processes, providing accurate predictions of the travel time and spread moving downstream in a natural stream [14]. This tool was applied only to the Mondego river, because the *DUFLOW* package has presented best performances and robustness during model calibration and validation procedures for this case study.

In 1998, DUFLOW is migrated to *WindowsNT* (DUFLOW 3.0) and has become a component of the *Duflow Modelling Studio* (DMS) [15].

The DMS can perform unsteady flow computations in networks of open water courses and can simulate more complex water quality processes. Furthermore, it includes a precipitation runoff module (RAM), from which the supply of rainfall to the surface flow, can be calculated and provides a ground water model (MODUFLOW), which is integrated with the DUFLOW model to simulate the interaction between ground water and surface water for watershed problem analysis. The DMS package was applied to Tagus river model development presented in this paper.

Prior their application, river models should be calibrated and verified with consistent datasets, based on experimental or field data of high quality, and covering a representative range of river flow values.

River dispersion models can also be applied in the development of early warning or alarm systems in order to determine the fate and the delay of a pollutant spill, accidentally discharged into a water body.

By the way, the application of *alarm models* allows the identification of the accident (location and occurrence) and the immediate limitation of its impacts by accurate control measures for water supply systems protection.

This issue has a special interest when applied to the desirable co-management of a trans-boundary river or reservoir.

3 Case studies

In order to cover comprehensive hydrodynamic regimes and river system characteristics, three case studies are presented in the following subsections.

3.1 Mondego river

Run-off from Urgeiriça uranium mine directly discharged to Pantanha streamlet, a tributary of Mondego river, has determined the interest of assessing the environmental impact on the receiving waters, considering the downstream presence of Carregal do Sal water supply impoundment.

3.1.1 Study area

The study area occupies the middle part of the Mondego river basin, located in the central region of Portugal. The drainage area is 6670 km^2 and the annual mean rainfall is between 1000 and 1200 mm. The river reach considered in this work begins downstream from the Caldas da Felgueira bridge and ends at the Tábua bridge (Fig.2), with a length of approximately 24 km.

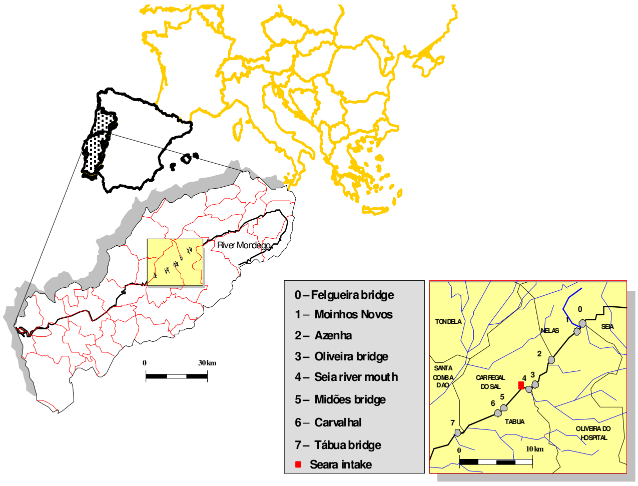

Fig.2: Mondego river basin and sampling stations

The river water is intensively used for hydroelectric power generation, domestic and industrial water supply and agricultural irrigation.

Seven sampling sites were considered, with site 0 (Caldas da Felgueira bridge) being the upstream tracer dye injection point. The only gauge station in the studded river reach is located in Nelas (before Aguieira dam).

The location of sampling stations was established according to their accessibility (bridges), mixing conditions, weirs location, logistics and human resources availability.

The flow regime of this river reach is strongly influenced by the Aguieira reservoir water level and by the fourteen weirs considered.

Water levels at Aguieira reservoir were recorded during the monitoring program. The flow discharge values considered for calculations were obtained from Nelas flow gauge station records.

3.1.2 Monitoring program

A monitoring program was carried out after injecting the tracer dye (*rhodamine WT*) in order to characterize *in situ* the transport and dispersion behaviour of the river under three different hydrodynamic regimes: flood (140 m^{3}s^{-1}), frequent (40 m^{3}s^{-1}) and dry-weather (0,74 m^{3}s^{-1}) flows.

Table 2 presents the information concerning all the tracer injections performed during the three sampling programs included in this study. Due to operational reasons, tracer dye was injected simultaneously in two distinct river sections.

Table 2: Synthesis of tracer injections (Mondego)

Injection	Date	Hour	Point	Flow (m^3/s)	Rhodamine mass (g)
1	89-12-09	8:20	Site 0	140	100
2	89-12-09	15:40	Site 3	144	200
3	89-12-10	8:00	Site 0	100	200
4	89-12-10	8:30	Site 5	110	400
1	90-06-15	7:32	Site 0	0.74	400
2	90-06-15	8:30	Site 3	0.74	200
1	90-11-09	7:40	Site 0	40	400
2	89-11-10	8:00	Site 3	29	400

Figure 3 shows the *rhodamine* cloud evolution for the first injection of the November monitoring program at Caldas da Felgueira bridge.

This site was selected immediately downstream from a weir, which increases water turbulence and mixing conditions, thus minimizing the tracer dye mixing length in this reach.

Fig.3: *Rhodamine* spreading after their injection (Mondego river at Caldas da Felgueira bridge)

3.1.3 Results and discussion

Table 3 compares the average velocity, travel time and dispersion results obtained from Mondego river model simulations with experimental tracer data.

Table 3: Mondego river model calibration results

MONITORING PROGRAM	REACH	AVERAGE VELOCITY (ms^{-1})		TRAVEL TIME (h)		DISPERSION COEFFICIENT (m^2s^{-1})		RECOVERED MASS
		EXPER.	DUFLOW	EXPER.	DUFLOW	EXPER.	DUFLOW	(%)
3 rd. (Nov.-90)	S1 – S2	0.526	Var.	2:37	2:35	14	10	57
	S2 – S3	0.497	Var.	2:41	2:41	51	45	56
	S3 – S5	0.473	Var.	3:21	3:19	37	35	55
	S1 – S3	0.511	Var.	5:18	5:16	34	-	-
	S1 – S5	0.497	Var.	8:38	8:35	35	-	-
1 st. (Dec.-89)	S1 – S2	1.105	Var.	1:14	1:14	52	40	62
	S2 – S3	0.949	Var.	1:24	1:24	61	70	62
	S1 – S3	1.023	Var.	2:38	2:38	58	-	-

Experimental longitudinal dispersion coefficients were calculated from concentration-time curves at consecutive sampling sites using the analytical solution of first order decay kinetics [16].

The good agreement between model results and experimental data can be supported by the range of correlation coefficients values calculated: 95% for sites 1 and 2; 98.5% for site 3, validating the longitudinal dispersion coefficients adopted for the Mondego river model calibration. Figure 4 shows the correlation between experimental concentration-time curves, the analytical solution values and the Mondego river models outputs (based on *Duflow* and *ADZ packages*), at sampling stations 2 and 3.

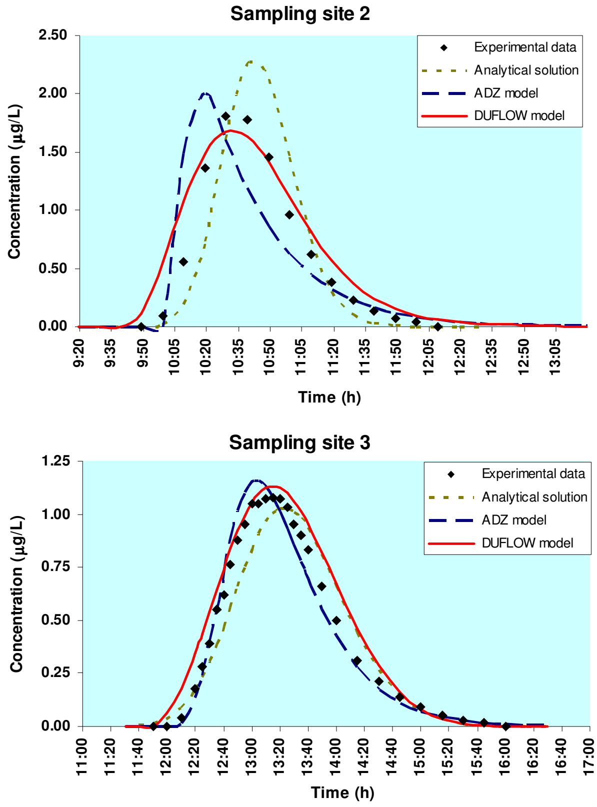

Fig.4: Mondego river models calibration with experimental data (S2 and S3 sampling sites)

A relatively better performance of the *DUFLOW* package can be inferred from these graphics. The Mondego river model was validated using experimental data from the December 89 monitoring program (first injection), under flood flow conditions. A good correlation with field data was also obtained, as supported by the correlation coefficient values depicted in Figure 5.

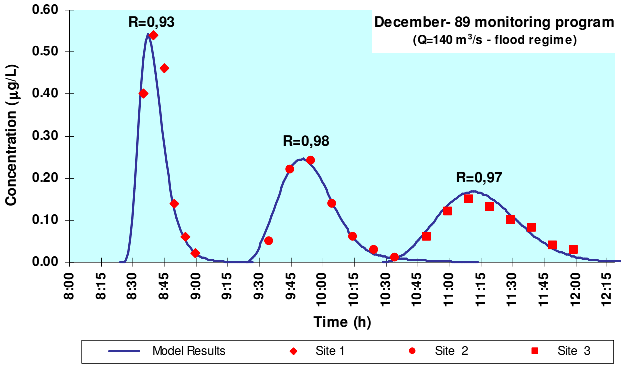

Fig.5: Mondego river model validation and correlation coefficients

In practice, river water dispersion characteristics can be evaluated from the peak concentration decrease with dye spread travel time variation at a downstream site [17]. After initial tracer and river water mixing, the ratio – peak concentration (Cp) / total injected tracer mass (M_{inj}) – decreases with a power function of travel time (Fig. 6).

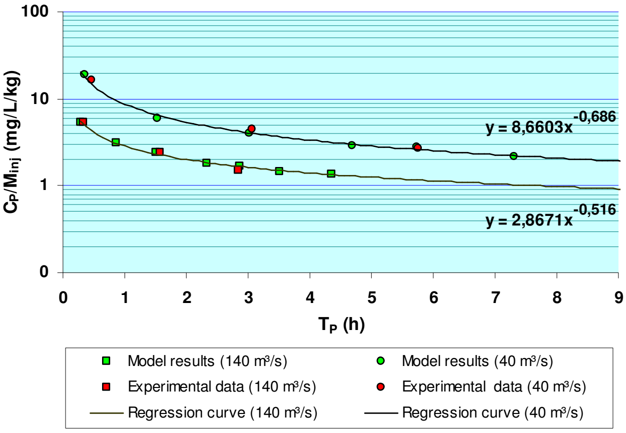

Fig. 6: Peak concentration variation with dye spread travel time (Mondego river)

3.2 Douro river (Miranda reservoir)

This case study has been designed with the purpose of assessing the environmental impact of an accidental pollutant discharge into the international reach of the Douro river, between the Castro dam (Spain) and the Miranda do Douro dam (Portugal).

3.2.1 Study area

The study area is located on a Douro river international reach at the north-eastern region of Portugal. This river reach begins downstream of the Castro reservoir and ends at the Miranda reservoir, with a length of approximately 13.5 km.

Four sampling sites were considered, with the site 0 (downstream Castro dam) being the upstream tracer dye injection point (Fig. 7).

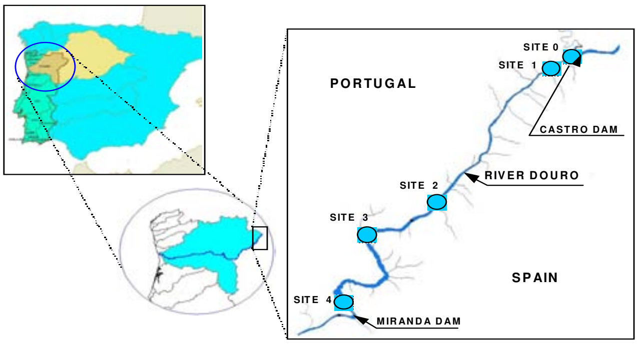

Fig.7: Douro river basin and sampling sites location

The flow regime of this river reach is strongly influenced by the discharged flow from the Castro dam, by the turbinated flow in the Miranda dam, and by the surface water level in the Miranda reservoir (Fig. 8).

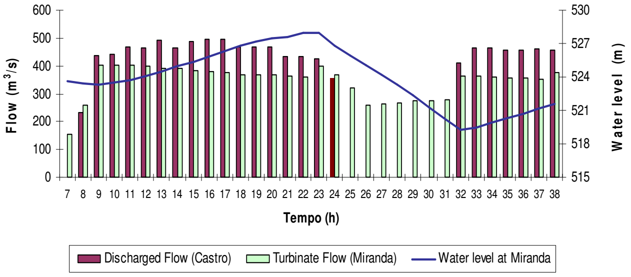

Fig.8: River Douro flow discharges and surface water level at the Miranda reservoir

3.2.2 Monitoring program

The upstream tracer dye injection point (site 0) was located downstream Castro dam, near the turbinated flow discharge point to get better mixing conditions.

Seven sampling campaigns were carried out using tracer injection (*rhodamine WT-20%* solution) to assess *in situ* the transport and dispersive behaviour in the reservoir water under flow rates ranging from 170 to 457 $m^3 \cdot s^{-1}$.

Average values of flow discharges at the Castro reservoir and mass of injected rhodamine were considered as the upstream boundary conditions. Surface water level at the Miranda reservoir was taken as the downstream boundary condition.

Four sampling sites were considered with the cross-sections depicted in Figure 9.

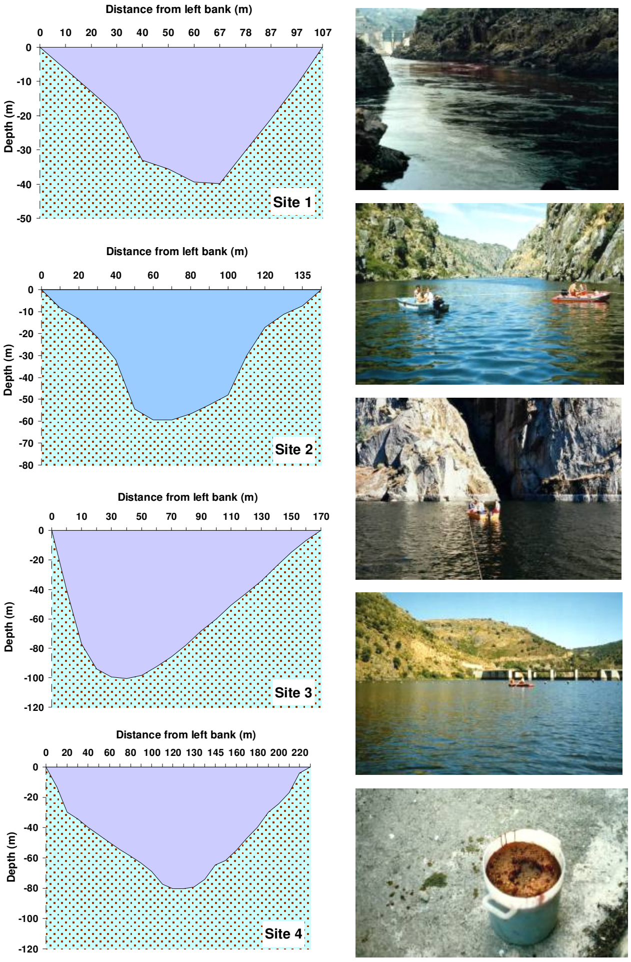

Fig.9: Sampling sites: bathymetry and characteristics

Water samples before the dye injection (blanks) were taken at all sampling points for determining the river reaches natural fluorescence.

Table 4 presents the information about all tracer injections obtained in the seven sampling campaigns scheduled for this study.

Table 4: Synthesis of tracer injections (Douro river)

Date	River flow (m^3/s)	Water level ** (m)	Rhodamine mass (kg)	Sampling section L	D	T
85-05-07/09	400	-	11.5 *	1-4	-	-
85-09-24/26	170	-	5	1-4	-	-
86-10-01/03	254	524	5	1-4	-	-
86-10-29/31	265	526	5	1-4	2-4	3, 4
87-04-08/10	457	525 - 522	5	1-4	3	2, 3
87-07-22/24	100	527 - 526	5	1-4	2	1, 2
87-11-18/20	352	525 - 524	5	1-4	3	1-3

Note: L = longitudinal ; D = on depth ; T = transversal
* - Rhodamine B ; ** - Miranda reservoir

Llateral mixing patterns of the tracer in the sampling site 2 are illustrated in Figure 10 [18].

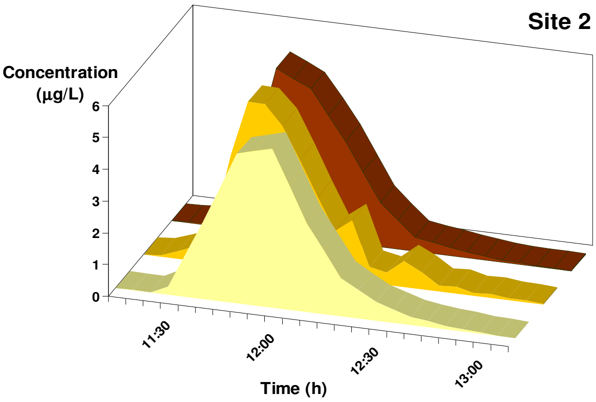

Fig.10: Transversal variation of tracer dye concentration (Miranda reservoir - site 2)

The time-concentration curves were obtained at 25, 50 and 75% of the river width, at that site, and no significant differences are observed among them.

Similar results were obtained at the other sites allowing the conclusion that mixing conditions were favourable to a rapid equalisation of concentrations in the transversal direction.

3.2.3 Results and discussion

The model calibration procedure (Fig. 11) included the adjustment of the friction bottom values and the longitudinal dispersion coefficients.

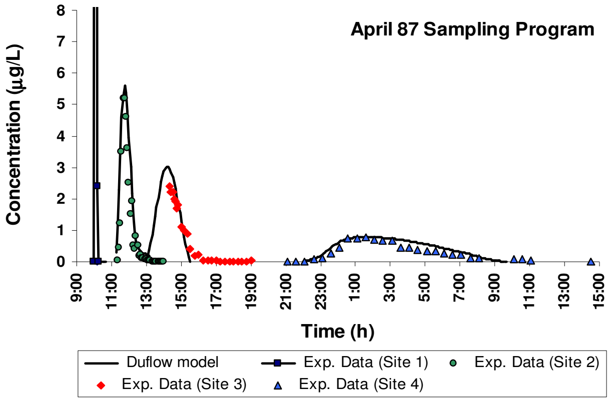

Fig.11: Douro river model calibration with field data

The model results show a good correlation with experimental data, accurately reproducing the tracer peak concentrations and the travel time between consecutive sampling stations.

The model was validated using other sampling data set (November 87), obtained under a different flow regime (Fig. 12).

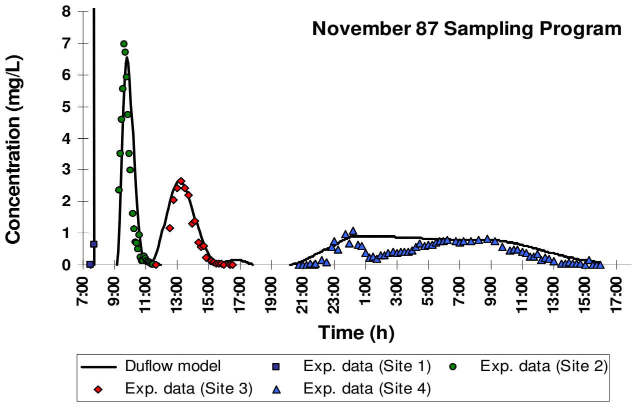

Fig.12: Douro river model validation with field data

Table 5 compares travel times and dispersion coefficients obtained from the fitting of Douro river model to the experimental data used for the calibration procedure.

Table 5: River Douro model calibration results

REACH	TRAVEL TIME (h)				LONG. DISPERSION COEFICIENT ($m^2.s^{-1}$)	
	APRIL 87		NOVEMBER 87		MODEL RESULTS	
	FIELD DATA	MODEL	FIELD DATA	MODEL	APRIL 87	NOVEMBER 87
Site 0 - Site 1	0:05	0:05	0.06	0:05	50	45
Site 1 - Site 2	1:45	1:45	2:10	2:20	30	20
Site 2 - Site 3	4:10	4:20	5:45	5:40	5	20
Site 3 - Site 4	15:35	15:45	16:45	17:30	2	2

The presented values show slight differences between experimental longitudinal dispersion coefficient values and those adopted for model calibration. This can indicate that a 2D-V modelling approach must be adopted in future works in order to improve the correlation between model results and experimental tracer data.

It was observed that the *ratio* between the tracer peak concentration (Cp) and the total mass of injected *rhodamine* varies with a negative power of the travel time (Fig. 13).

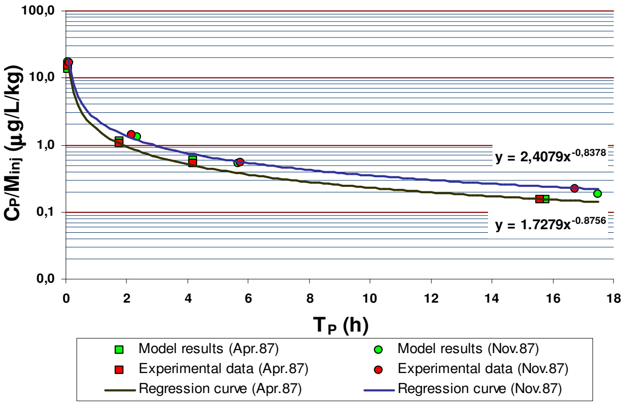

Fig.13: Peak concentration variation with dye spread travel time (Miranda reservoir)

This exponent is a constant (ranging from 0.84 to 0.86, depending on the river flow rate and reach dispersion characteristics

3.3 Tagus river (Belver reservoir)

This case study has been planned with the purpose of evaluating the environmental impact of accidental radioactive cooling water discharges from a nuclear power plant, upstream, in Spain, in the international reach of Tagus river, between the Cedillo dam (Spain) and the Belver dam (Portugal).

3.3.1 Study area

The study area selected for this work is located on a Tagus river national reach. This river reach begins downstream from the Fratel reservoir (Portugal) and ends at the Belver dam, with a length of approximately 9.2 km (Fig. 14).

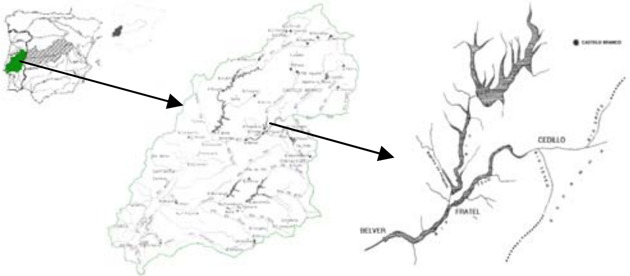

Fig.14: Tagus river basin and the studied reach

Four sampling stations were considered (S8, S9, S10 and S10a, with the site 8 (downstream from the Fratel dam) being the upstream tracer dye injection point.

The flow regime of this river reach is strongly influenced by the Fratel dam discharged flows and also by the turbinated flows and the surface water levels at the Belver dam.

3.3.2 Monitoring program

Four sampling campaigns were carried out using tracer injection (*rhodamine WT*) to characterize *in situ* of the transport and dispersive behaviour in reservoir water under flow ranges from 23 to 300 $m^3 \cdot s^{-1}$. Table 6 presents the information about all tracer injections made in this study.

Table 6: Synthesis of tracer injections (Tagus river)

Data	Injection point	Sampling sites	River flow (m^3/s)	Water level (m)	Tracer dye mass (L)
1991-06-08/11	Cedillo Fratel	S2, S3, S4, S9, S10, S11	150	73.0 (Fratel) 48.0 (Belver)	10 5
1991-11-23/26	Cedillo Fratel	S2, S3, S4, S9, S10, S11, S12	300	72.7 (Fratel) 45.9 (Belver)	10 10
1992-04-12/15	Cedillo	S1a, S2, S2a, S3, S4	148	72.75 (Fratel)	20
03-11-1992	Fratel	S8, S9*, S10, S10a	23	47.0 (Belver)	5

The model conceptualization was based on DMS scheme and Google Earth skills. The segmentation of Fratel and Belver reservoir adopted for Tagus river modelling is presented in Figure 15.

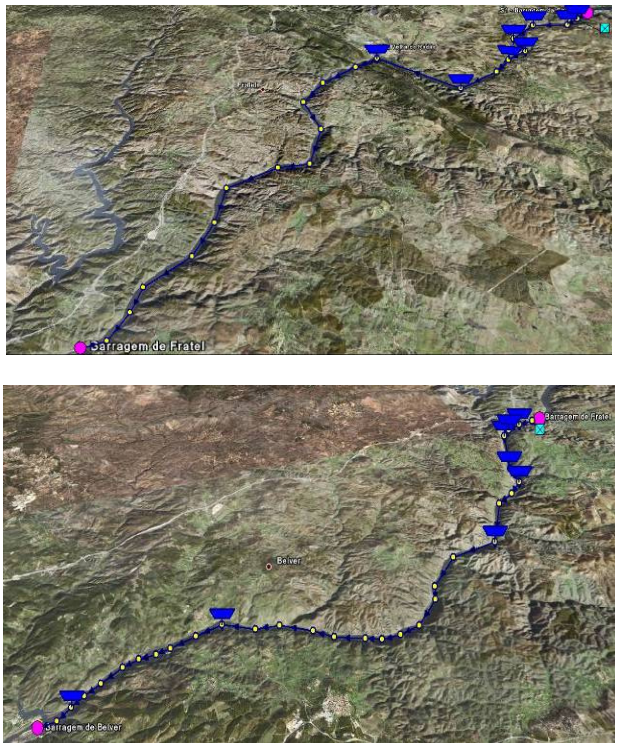

Fig.15: Tagus river model spatial schematization

Average values of flow discharges at the Fratel reservoir and mass of injected *rhodamine WT* were considered as the upstream boundary conditions. Surface water level at the Belver reservoir was taken as the downstream boundary condition.

3.3.3 Results and discussion

The model calibration procedure included the adjustment of the friction bottom value in each reach and longitudinal dispersion coefficients obtained in the sampling program of November 92.

Table 7 compares travel times and dispersion coefficients obtained from the Tagus river model outputs with experimental tracer data.

Table 7: Tagus river model calibration results

Program	Reach	Average velocity ($m.s^{-1}$)		Travel time (h)		Long. dispersion coeficient (m^2/s)	
		Exper.	Model	Exper.	Model	Exper.	Model
4th (Nov. 92)	**S8 – S8a**	0.156	0.1563	1:00	0:50	14.8	5
	S8a – S9*	0.126	0.126	1:25	1:16	7.5	9
	S9* – S9a	0.129	0.1291	2:43	2:50	7.3	8
	S9a – S10	0.111	0.111	2:45	3:14	11.4	8

The model was validated using other field sampling data (November 91), obtained under a different flow regime (Fig. 16).

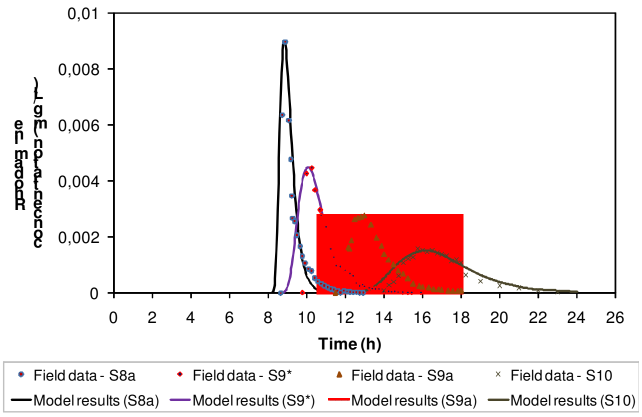

Fig.16: Tagus river model validation with field data

After the initial tracer and river water mixing, one can observe that the *ratio* – peak concentration (C_p) / total injected tracer mass (M_{inj}) – decreases with a power function of the travel time. The exponent is a constant in the range 0.77 - 1.23, depending on the river reach dispersion characteristics and the hydrodynamic regime ((Fig. 17).

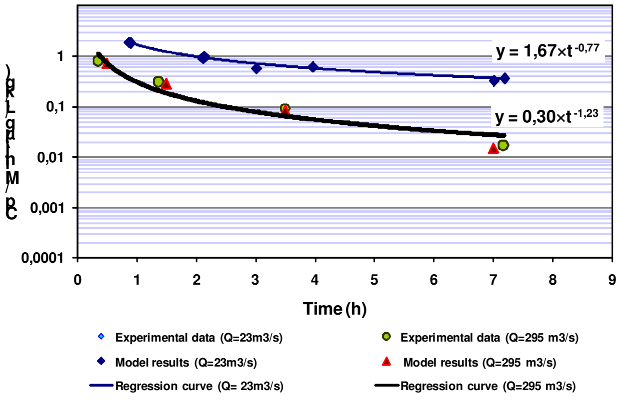

Fig.17: Peak concentration variation with dye spread travel time (Belver reservoir)

The exponent values range calculated in this case study is very high, greater than the values observed on the Miranda reservoir study, indicating that 2D or 3D models are probably more accurate and should be applied to this river system in order to improve the correlation between model predictions and tracer experimental data sets. This models can perform a better understand of the transversal and vertical mixing contributes to the monitored dispersive behaviour of this water body.

4 Conclusions

This work presents the results of longitudinal dispersion coefficients estimates and proposes mathematical equations for peak concentration decrease with dye spread travel time, based on tracer experiments that have been performed in several reaches of three different Portuguese river systems.

Mathematical modelling appears to be a powerful tool to solve pollutant transport problems in river systems with longitudinal dispersion behaviour similar to the case studies presented, even under different flow regimes.

For the Miranda and the Belver reservoirs further developments must be done to simulate vertical and transversal dispersion processes in order to improve the model results correlation with the experimental tracer data and to mitigate some prediction uncertainty.

The *DMS package* is a useful tool to develop accurate river models and to simulate pollutants transport in water bodies with different dispersive characteristics. In general, models results showed a satisfactory agreement with experimental data, allowing a reasonable support for impact assessment of different pollutant load scenarios in the river water quality.

This procedure is of paramount interest in river basin management strategy for defining early warning or alarm systems, minimizing the effects from accidental pollutant spills, and to improve water sources protection practices.

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