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River, Southwestern France

By:
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Flux of suspended particulate matter and cadmium in the Lot River, southwestern France.

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Management Perspective

- A research team consisting of scientists from the University of Bordeaux and the National Water Research Institute studied the dynamics of cadmium pollution in the Lot River system, southwestern France. Cadmium poses a real threat to valuable oyster beds located downstream near the mouth of the Gironde Estuary.

- The Cd contamination source is an abandoned zinc ore refinery located in the upper reaches of the Lot. Now, large amounts of Cd accumulated in local bottom sediments are being mobilized and transported by the river to the estuary.

- The purpose of the study was to find out the rate of Cd transport and the likely duration of contaminant input. Using innovative methodology, the annual Cd flux calculated ranged from 1.3 $t.y^{-1}$ (1990) to 17.5 $t.y^{-1}$ (1992).

- Mobilization and transport are then expected to purge the sediments of their Cd stock in 13 to 76 years; average flow values for the past 6 years would result in removal of Cd in less than 27 years.

- The study therefore provides a method for estimating the duration of an in-place contaminant problem; this is important information for decision-making on which remedial options are most appropriate for riverine situations such as the St. Lawrence River.

Abstract

The source of present-day cadmium contamination in the Gironde Estuary is an abandoned industrial facility once used for refining zinc ore, located in the upper reaches of the Lot River basin. In quantifying the fluxes of cadmium contamination in the river, a considerable effort had to be devoted to methodological development. A key factor in this methodological scheme was the measurement of fluxes of suspended particulate matter (SPM) for various flow regimes during the year. Although the flux of Cd is the dissolved state is important (it was found to be fairly constant at around 1 $t.y^{-1}$), most of the Cd is associated with the SPM (up to 13 $t.y^{-1}$). Another key factor was the application of an automatic pump sampler which collected an integrated daily sample from the main flow channel. The large set of measurements of turbidity (measured at Temple-sur-Lot) and volumetric discharge at monitoring sites located at Villeneuve-sur-Lot, allowed quantitative comparison of two techniques of calculating SPM flux. Of the two, the stochastic technique was judged best suited to automatic sampler data and also provided a better estimation of Cd flux. The other, the deterministic technique, was useful in yielding a meaningful relationship between river discharge and SPM that covered 73% of the cases measured, thus enabling reliable estimation of the major part of SPM and associated Cd (particulate Cd) fluxes from discharge data alone. However, it consistently overestimated Cd flux during drier years. Nevertheless, these drier years are characterized by a minimum of SPM and particulate Cd fluxes, so the global consequence of the overestimation is very limited. Using the stochastic technique, values for particulate Cd flux were calculated which ranged from 1.3 $t.y^{-1}$ in 1990 to 17.5 $t.y^{-1}$ in 1992. Estimations of total Cd fluxes contributed by the Lot River to such a level of precision are deemed critical in directing clean-up efforts in the basin and estuary as a whole.

<u>Key words:</u> Cadmium pollution, SPM measurement, fluvial processes, southwest France

INTRODUCTION

Cadmium contamination of natural oyster beds at the mouth of the Gironde estuary is well-known (1). The fluvial origin of the cadmium, combined with the normal estuarine processes that have led to this pollution, has been already brought out in various research efforts on the behaviour of metals in the Gironde (2, 3). In the framework of a study involving all the watersheds draining into the Dordogne and the Garonne Rivers (4, 5), it was shown that the cadmium reaching the Gironde originated to a large degree (90% in 1987) from the industrial region of Decazeville in the upper Lot River basin (Fig. 1), initially via a small tributary, the Riou-Mort. This stream receives surface runoff water from unstabilized and open mine-tailings mounds and also from industrial discharges from the Vielle Montagne plant. The chronic metal pollution of the Lot was recognized since 1971 and, following a large accidental discharge of cadmium in July 1986 (6), a number of remediation works were carried out primarily to isolate the tailings mounds from the local water table and nearby streams, and also to reduce to a minimum the direct discharges of the plant. These measures resulted in a considerable point-source reduction of the flux of cadmium from 40 $kg.day^{-1}$ in 1986 to 3 $kg.day^{-1}$ in 1991, according to plant estimates. Despite this significant source reduction, the contaminated sediments accumulated over decades on the bed of the Lot remain susceptible to maintaining high fluxes of cadmium through resuspension or through diffusion to the overlying water column.

The paper presents a detailed study of cadmium flux, discharged via the Lot into the Garonne River. The study also examines the methodological requirements for long-term monitoring, especially the establishment of a rigorous sampling protocol to ensure as much as possible the continuity of the measurements involved. The principal challenges presented to the study were those related to the establishment of a mass balance for the suspended particulate matter (SPM). The accuracy of this budget is of great importance because the SPM represents the main dispersal vectors for cadmium and other trace metals, especially zinc.

FIELD AND LABORATORY METHODOLOGY

During the period 1990-1991, all SPM and water samples were collected in the lower Lot River at Clairac (Fig. 1). Beginning in 1992, the sample site has to be moved 8 km upstream to Temple-sur-Lot because of the construction of a lock near Clairac. Flow parameters, measured slightly downstream at the station at Villeneuve-sur-Lot, were provided by the Service Hydrologique et des Milieux Aquatiques de la Direction Régionale de l'Environnement Midi-Pyrénées (Hydrological Service of the Department of the Environment, Midi-Pyrenees Region). The precision of discharge measurements below 40 $m^3.s^{-1}$ is not ideal, but fortunately, this situation involves only the low-flow discharges when the SPM fluxes are also very low (7).

The SPM concentration was determined by filtration (Whatman GF/F filter, porosity - 0.47 µm) followed by weighing of 0.5 to 1 litre aliquots of water. Relative error for these measurements was consistently less than 1.5%.

Sampling frequency was initially monthly, with a more detailed monitoring during the main high-flow periods (2 or 3 samplings per day during each of these periods). Following the first two mass-balance calculations carried out in 1990 and 1991, it became apparent that a better understanding of SPM flux required a much denser sampling of turbidity, especially during the high-flow periods. Starting in 1992, manual sampling frequency was changed to daily; since 1993, however, an automated sampling system was installed which could take an integrated sample every 90 minutes. The traditional equal-time-step sampling strategy was selected as it was deemed easier to implement than the strategy of sampling proportional to water volumes or SPM quantities. The sampling time-step was set sufficiently low to minimize variations in fluid or solid discharge during this time-interval. The automatic sampler (Bühler, model PB-MOS) provided an average daily sample consisting of the sum of 16 individual samples collected in a single bottle. The volumes sampled were determined by assuming that the SPM in the volume of water entrained by the pump was representative of the SPM level in the river, allowing for the rated efficiency of the pump.

The samples were collected in the main flow channel at a depth of 2 metres. Tests of representativeness since the sample is collected over a period of about 1 hour. The SPM in the sample was separated initially by centrifugation (Westfalia separator, hourly capacity of about 135 L). The particles, subjected to an acceleration of 12,000 G, were recovered at 95% efficiency without significant metallic contamination with respect to the target elements (9). The water exiting the separator was then filtered through a stack of 293 mm diameter filters (0.47 µm Micropore polycarbonate) to recover the finest particles. The use of these two techniques in sequence allows an almost complete recovery of the SPM; minimum recovery level was on the order of 98%. After recovery, the two separates were dried, weighed, and homogenized prior to analysis and turbidity determination of the water sampled. The SPM samples were digested in a Teflon bomb using HNO_3 and HCl according to the method of Loring (8).

The elemental analysis was carried out using an Electrothermal atomic absorption spectrometer (ETAAS). The accuracy and precision of the measurements were tested on the basis of an intercalibration exercise using ICES international standards (8). In view of the consistently high metal concentration values, the analysis for dissolved metals was carried out directly on the filtered water.

RESULTS

Water discharge / turbidity relationships

Two above-average years (1992 and 1994) and four below-average years (1989, 1990, 1991, and 1993) can be identified compared to the mean annual discharge (176 $m^3.s^{-1}$) measured at Villeneuve-sur-Lot for the 1973-1988 period (Figure 2). In fact, the values for 1989, 1990, and 1991, though low, do not accurately reflect the severity of the drought that took place in those years because the Lot is regulated to maintain discharge levels during low-flow periods by releasing water out of the dams in upper reaches of the river. In these years, the number of days of high-flow conditions when the discharge was above 400 $m^3.s^{-1}$ (11 days in 1990 and 2 in 1991) was very low compared to the average of 32 days during the past 21 years (Table 1).

Measured SPM loads are presented Figure 3 as a function of daily water discharge for all the samples collected between 1990 and 1994 (n = 1927 samples). In general, the values show a high level of scatter. Nevertheless, the presence of several distinct groupings of points allows the identification of a significant discharge / turbidity relationship for a large proportion of the samples. The three groupings thus distinguished are described below.

The first group (4.8 % of the total of samples) corresponds to the measurements taken in high surface-runoff periods at the site of the sampling station (Fig. 3; solid stars) when precipitation was greater than 20 mm per day, i.e. 92 samples. The high turbidity events that were noted at these times are probably due to "bursts" of SPM associated with surface runoff in the vicinity of the station. Such runoff effects are magnified by the fact that the soils of the Lot valley, an important agricultural region, are characterized by a high level of erodibility (10, 11).

The second group (22.0 % of the total of samples) is represented by samples collected during low stream-flow periods (Fig. 3, open stars) where the discharge values are below 40 $m^3.s^{-1}$ and the particulate organic carbon concentration is generally above 7%. The SPM concentration for these samples is relatively high. These measurements correspond to the spring and summer surveys and coincided with intense autochthonous vegetation growth (plankton blooms, macrophytes, sundry aquatic vegetation). This enrichment in organic carbon, followed by eutrophication of bottom waters (12) appears to be in part due to anthropogenic activities over the past decades. The construction of dams tends to homogenize the river flow and to encourage a peak-flattening in the high-flow periods. The creation of constant-level zones also leads to a considerable change in the physico-chemical quality of the water (13) which favours phytoplanktonic growth in the stagnant-water areas (14, 15).

The third group (Fig 3, solid dots) is formed by the remaining points. It corresponds to the most frequent situation (1412 samples, i.e. approximately 73% of the initial population). For this third group a clear relationship between SPM concentration (C_{SPM}) and discharge (Q) appears. By using the general expression recommended by Meybeck et al. (16), the relationship can be defined by equation (1):

$$C_{SPM} = a\,(Q_{discharge})^b \qquad (1)$$

where the calculated values for the constants are: $a = 4 \times 10^{-4}$ and $b = 1.92$. The correlation coefficient ($r = 0.84$) thus obtained between SPM and discharge is significant (95% confidence level for the correlation coefficient estimate), but the relationship is only representative of 73% of the samples, including those with the highest SPM concentrations. The significant correlation between SPM and discharge is demonstrated further in Figure 4 for high-flow periods, such as during the floods of June 1992. Taking into account the fact that these 1412 samples represent the highest concentrations, the resulting SPM fluxes then correspond to as much as 84% of the total fluxes. This means that the uncertainty related to the SPM flux calculation is only about 16%.

Calculation of SPM flux

Calculation of fluxes of SPM and other pollutants has been discussed at length by many workers. In this paper we consider in particular the work of Rakoczi (17), Meade and Parker (18) and Meade and Stevens (19). Two principal techniques have been used to calculate SPM fluxes.

In the first method, the so-called stochastic method, SPM concentration measured during each survey can be considered representative of concentrations over the time-interval between two surveys. This method is therefore well-suited to the type of data provided by the automatic sampler, and allows calculation of daily flux values using the daily mean SPM concentration (C_{SPM}) from 16 daily samples, combined with the mean daily discharge (Q). The annual SPM flux (F_{SPM} is the sum over one year of the product:

$$F_{SPM} = \Sigma^{365} Q \cdot C_{SPM} \qquad (2)$$

The river gauging system used by the Midi-Pyrenees Hydrological Service has a theoretical measurement error for the fluid discharge of less than 5%; the determination of SPM concentrations from the samples is carried out with a relative uncertainty well below 1%. The high sampling frequency (every 90 minutes) employed since 1992 allows the error associated with the temporal representativeness of the sample to be limited to 10%. In this way, it is possible to estimate that a combined relative error for the SPM flux calculation would be approximately 15%.

The second approach, the so-called deterministic technique, consists of applying the relationship which exists between SPM and water discharge. The fluxes exported by the Lot were calculated with the aid of the relationship presented above (Equation 1) between water discharge and SPM concentration. In the present case, given the fact that the relationship was found to be representative for only 73% of the samples, but

84% of the SPM flux, the uncertainty related to the SPM flux calculation is in the vicinity of 16% using this technique.

The results obtained from the two techniques are compared in Table 2. A noticeable difference between the two methods appears for 1990 and 1991, years in which discharge values were low. In such conditions, the discharge/SPM concentration relationship was not uniquely dependent on the dynamics of the river, but also on plankton-related developments. Conversely, the other years show that, when SPM flux is more than 0.3 $Mt.y^{-1}$, the results are virtually identical, and the differences do not exceed 20%.

Calculation of cadmium flux

The total flux of Cd comprises the sum of dissolved cadmium and that of cadmium associated with suspended matter. The fluxes of dissolved cadmium ($F_{Cd,d}$) are calculated using the product of mean concentration and discharge (Q_i):

$$F_{Cd,d} = Q \,.\, (\sum Q_i \,.\, C_i / \sum Q_i) \qquad (3)$$

where Q = mean discharge, Q_i = instantaneous discharge , and C_i = the instantaneous concentration.

The results (Table 3) show that dissolved cadmium fluxes remain relatively constant, and that there is no clear relationship between fluid discharge (Table 2) and dissolved cadmium flux (Table 3). Calculation of particulate cadmium flux ($F_{Cd,p}$) as a function of the cadmium concentration ($C_{Cd,p}$) has been made according to the method recommended by Meybeck et al. (16) for the case where:

- particulate cadmium fluxes are more dependent on SPM concentration than on that of cadmium;
- temporal variations in the concentration of particulate cadmium are small and thus do not necessitate too large a number of samples;
- a reasonably good knowledge of SPM flux (F_{SPM}) is available.

If these three conditions are met, the flux of particulate cadmium can be calculated from the following equation:

$$F_{Cd,p} = \sum F_{SPM} \cdot C_{Cd,p} \qquad (4)$$

Tables 4 and 5 show the fluxes of cadmium calculated using the two techniques of SPM flux calculation described above. It is obvious that the calculation technique for cadmium fluxes using the SPM values obtained by the deterministic technique, i.e. relationship linking SPM flux to discharge (Table 5), yields results that are significantly different. This difference may be explained by the fact that the relationship linking discharge and SPM concentration is represented only by 73% of the samples, and notably does not include those covering the periods of low flow. In particular, a considerable over-estimation is noted in the cadmium flux during the drier years. The stochastic technique is therefore favoured for the estimation of cadmium flux.

Total cadmium fluxes calculated using this technique are presented in Table 6. The relationship between mean annual discharge and cadmium flux is particularly clear in the case of the particulate phase. The dry years (1990 and 1991) are associated with low cadmium fluxes, while the wetter years (1992 and 1994) show much higher cadmium flux values.

DISCUSSION

The characterization of fluvial systems has often been investigated on the basis of discharge / SPM relationships in the watercourses studied. In many cases, the correlation obtained is inconclusive and it is generally admitted (20) that this result is due in part to the complexity of the processes controlling suspended sediment loadings and in part to measurement uncertainties (difficulties in sampling during the high flow periods, insufficient sampling stations, sample representativeness, etc.). In this regard, the Lot case study may be instructive. The methodology used at the beginning of the study showed the necessity for an increase in the frequency of measurements for all discharge regimes. The discharge : turbidity relationship was monitored (Fig. 4) during various types of flow regimes in the Lot: spring freshets (June 1992, April - May 1993, April - May 1994) as well as the fall / winter high-flow periods (October - November, 1992, December 1993 - January and February 1994). In all cases, the relative variation of the discharge values within a 90-minute period (between two automatic samplings) was less than 3%, i.e. virtually the same order of magnitude as the gauge measurement precision (5%). Similarly, the relative variation of the SPM concentrations within a similar 90 minute period was on the order of 8%, in other words, of the same order of magnitude as the error due to the representativeness of the sample (10). The sampling strategy can therefore be judged as acceptable, even if it is generally redundant, particularly during low-flow periods.

The use of a rigorous methodology for the sampling surveys, when combined with the selective approach in interpreting the data, allowed us to determine the flux of SPM according to two techniques:

- the stochastic technique is based on a high sampling frequency, and, appears to be the better choice for the determination of total material flux. Its precision is on the order of 15%.

- the other (deterministic) technique makes use of the relationship between fluid discharge and solid load in attempting to explain the cases that fall outside this relationship. This method is able to explain no more than 73% of the sample cases. The uncertainty is admittedly greater, but the technique has the advantage of being able to provide, from the discharge values alone, a less precise, but more rapid, estimate of the solid particle flux exported.

Such results therefore are of considerable interest as they offer possibilities for modelling fluvial transport in the Lot River in particular, and in a more global sense. A total material flux value at an annual scale depends necessarily on a relatively high sampling frequency. The SPM flux balance thus obtained appears to be sufficiently reliable to serve as a basis for calculating the flux of pollutants in particulate form carried by the Lot River en route to the Garonne River.

The methodology used for the monitoring of long-term SPM flux was the object of a preliminary study between the years 1986 and 1989. This study was instrumental in the design of a global strategy that was well adapted to the hydrological regime of a river whose flow is regulated by a large number (62) of dams and flow-retention structures. The river nevertheless exhibited periods of high discharge and variations in the nature of the suspended load. In order to obtain an accurate knowledge of the solid discharge, and to monitor the SPM concentrations with time, the development of a systematic monitoring plan was necessary . Automation of the sampling process was deemed necessary and resulted in a sampling frequency level that could be termed redundant, especially in periods of low flow. The sampling plan for the dissolved metal measurements required working under ultra-clean conditions (i.e. design of equipment for specific recovery, decontamination of sampling equipment, mobile clean laboratory equipped with a laminar flow hood, etc.). Similarly, the recovery of large quantities of SPM required heavy-duty, but sensitive, field equipment.

The large number of measurements carried out allowed us to extract a meaningful relationship between SPM concentration and fluid discharge. However, this relationship still is not representative enough of the whole sample data set to allow use of SPM measurements alone for calculation of metal flux. The choice of calculation method for cadmium flux in particulate and in dissolved form was dictated by the sampling strategy as recommended by Meybeck (21) and Meybeck et al. (16). The results showed a strong but complex relationship linking fluid flux with that of particulate cadmium.

Finally, the accurate understanding of the fate of cadmium-pollution fluxes exported to the Garonne River could permit us to monitor the fate of the cadmium presently stockpiled in the bottom sediments of the Lot river.

By using the above-described methodology and taking into account the previously-estimated stock of cadmium contamination in the Lot River sediments (7), it is now possible to predict the evolution of this stock with time. If the maximum annual fluxes measured during the wettest year, 1992 (18.2 $t.y^{-1}$) are considered, then the complete removal of the Cd would be achieved in about 13 years. For the drier years, the much lower annual fluxes calculated (2.3 $t.y^{-1}$ in 1991) mean that a complete purging of the Cd would take as long as 76 years. Using the mean value for the past 6 years of measurements (i.e. 7 $t.y^{-1}$), natural Cd background levels would be reached within less than 27 years dating from 1991, i.e. before 2018. This knowledge could be useful eventually in the progressive reduction of the present polluted state of the Lot River, and directing clean-up efforts throughout the basin.

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Table 1. Number of days with flows greater than 400 $m^3.s^{-1}$.

1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994
8	26	15	45	38	52	66	46	39	39	29	22	13	28	1	71	15	11	2	40	14	85

Table 2. Annual volume discharge and SPM flux calculated using two different methods.

Years	1973-1979	1990	1991	1992	1993	1994
Means of annual water discharge m^{-3}. s^{-1}	174	129	107	198	128	243
Annual SPM flux (stochastic method) Mt. y^{-1}		0.045	0.044	0.670	0.280	0.842
Annual SPM flux (deterministic method) Mt. y^{-1}		0.161	0.109	0.542	0.260	0.822

Table 3. Annual flux of dissoved Cd for the Lot River, 1990-1994.

Years	1990	1991	1992	1993	1994
Number of analyses	15	20	20	18	28
Standard deviation $\sigma_{(n)}$, ng.L^{-1}	64	48	37	41	35
Means of Cd concentrations, ng.L^{-1}	295	261	112	121	119
Annual flows of dissolved Cd t. y^{1}	1.2±0.2	0.9±0.2	0.7±0.2	0.5±0.2	0.9±0.1

Table 6. Annual flux of total Cd (stochastic method) and mean annual water discharge for the Lot River, 1990-1994.

Lot	1990	1991	1992	1993	1994
Annual total Cd t. y^{-1}	2.5±0.5	2.3±0.4	18.2±2.9	4.4±1.3	14.8±3.2
Mean annual water discharge $m^3.s^{-1}$	129	107	198	128	243

FIGURE CAPTIONS

Figure 1 Location map of the Lot River basin, southwestern France, showing place names and sampling sites.

Figure 2 Histograms of annual volumetric discharge measurements taken at Villeneuve-sur-Lot (1973-1994). Two above-average years (1992 and 1994) and four below-average years (1989-1991, 1993) are clearly identified.

Figure 3 Plot of 1927 daily measurements of SPM vs. corresponding volumetric discharge in the Lot River (1990 -1994). Solid stars (4%) represent measurements collected during periods of high surface runoff; open stars (22%) represent those with high organic content; solid dots (73%) represent those collected during more "normal" conditions.

Figure 4 Plot of SPM vs. discharge for the Lot River during the floods of June 1-21, 1992.

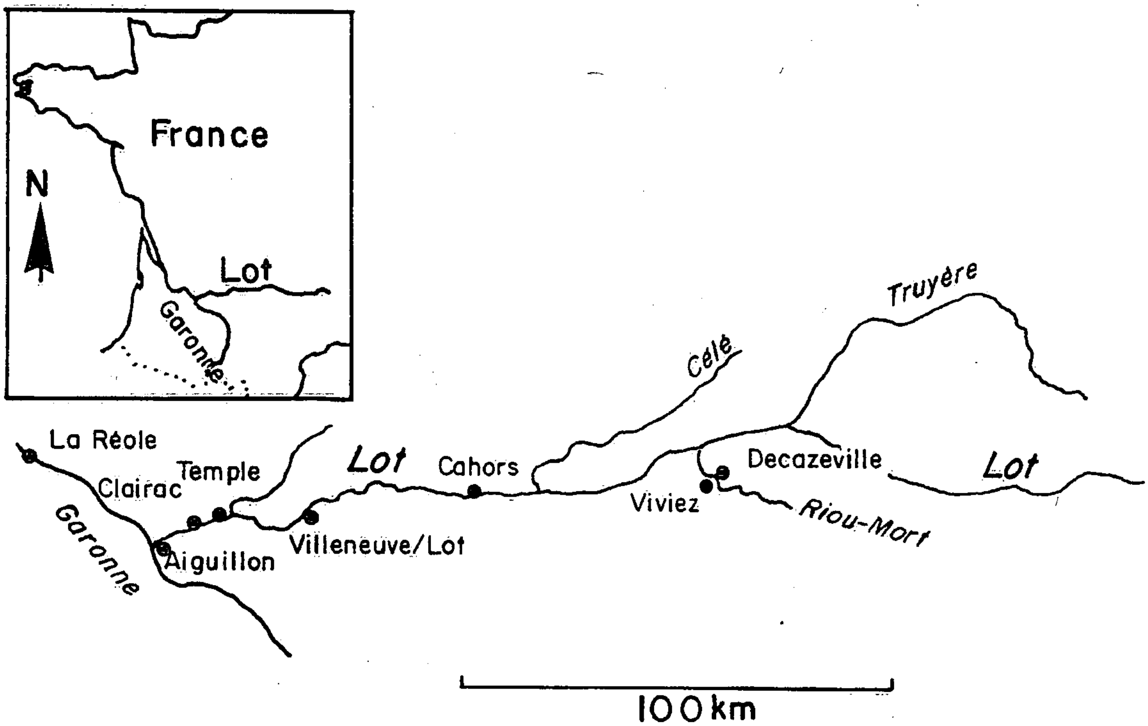
France
N
Lot
Garonne
La Réole
Clairac
Temple
Aiguillon
Garonne
Lot
Villeneuve/Lot
Cahors
Célé
Truyère
Decazeville
Viviez
Riou-Mort
Lot
100 km

Fig 1

Fig. 1

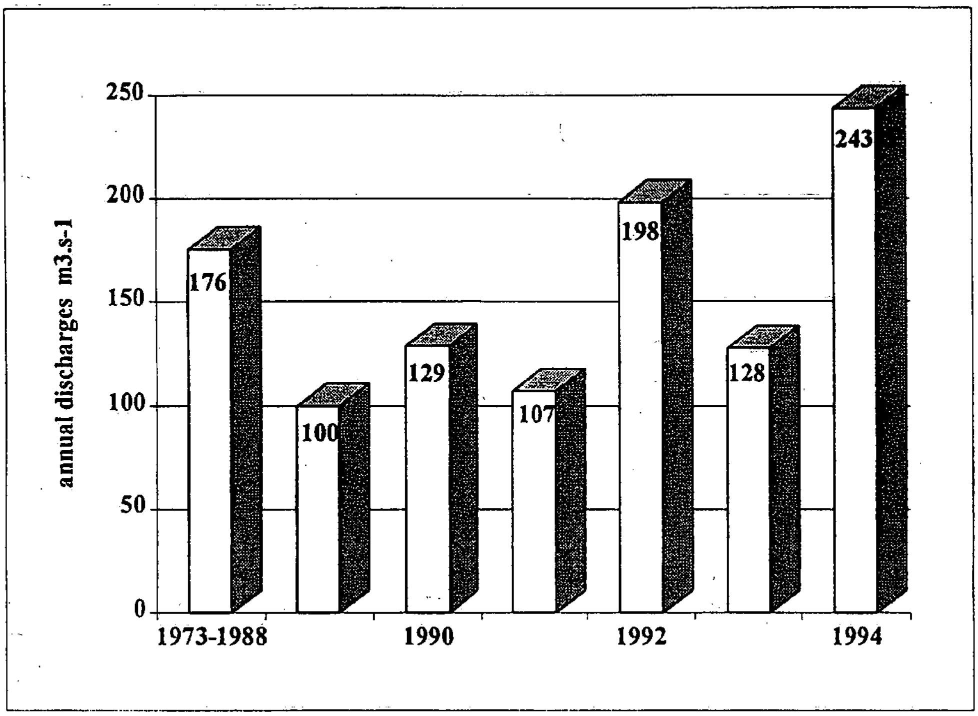
250
200
150
100
50
0
annual discharges m3.s-1
176
100
129
107
198
128
243
1973-1988
1990
1992
1994

Fig. 2

Fig. 2

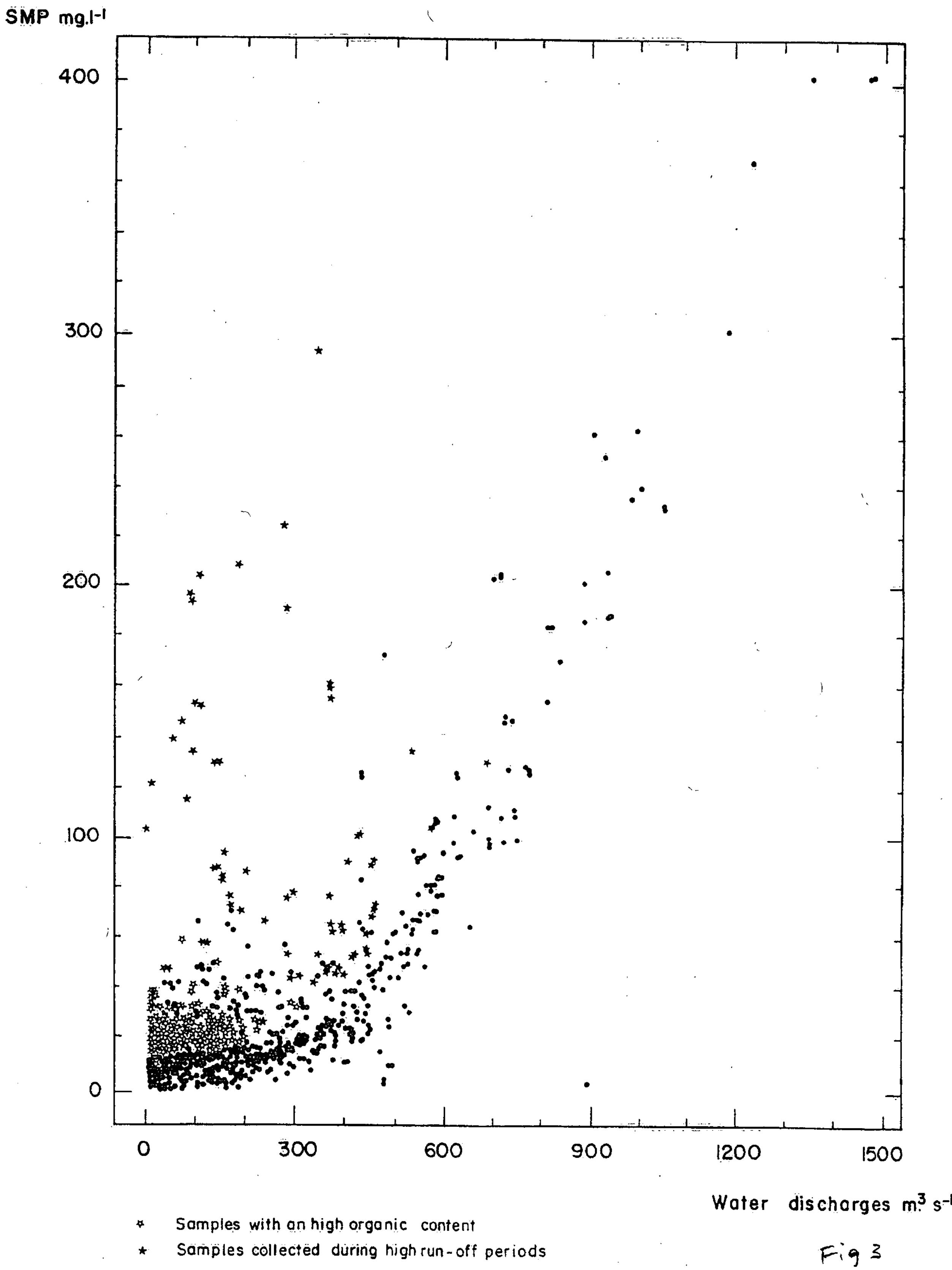
SMP mg.l-1
400
300
200
100
0
0
300
600
900
1200
1500
Water discharges m3 s-1
Samples with an high organic content
Samples collected during high run-off periods

Fig 3

Fig. 3

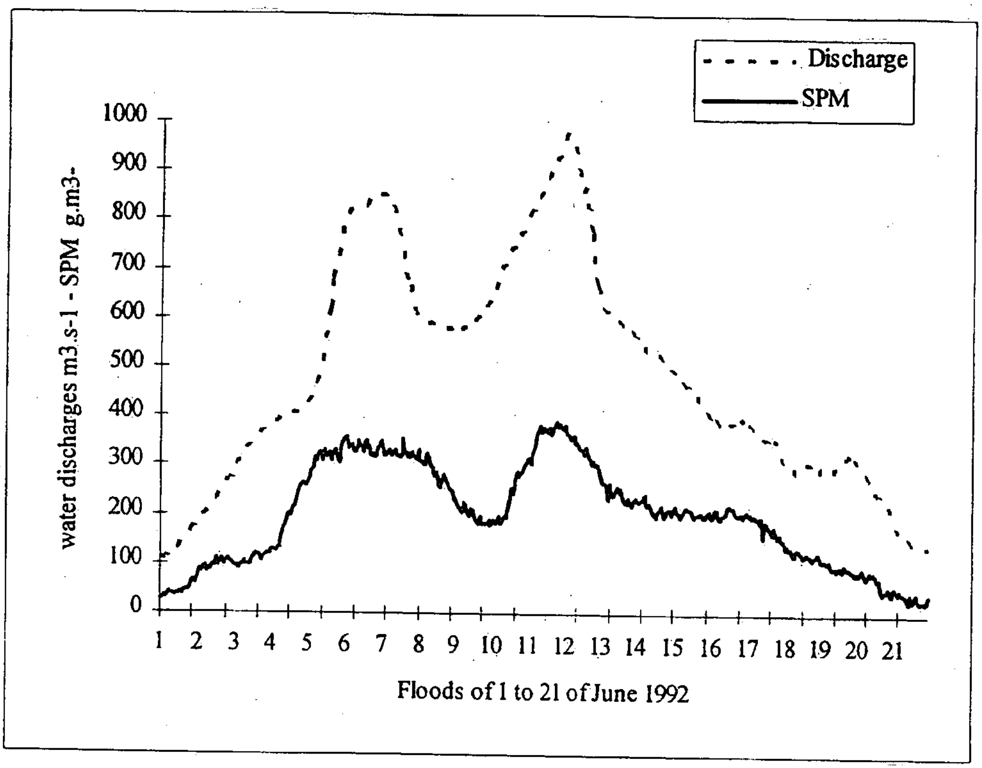
Discharge
SPM
1000
900
800
700
600
500
400
300
200
100
0
water discharges m3.s-1 - SPM g.m3-
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21
Floods of 1 to 21 of June 1992

Fig. 4

Fig. 4

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