**Effects of flushing flows on the transport of mercury-polluted particulate matter
from the Flix Reservoir to the Ebro Estuary**

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Abstract

Polluted sediments retained in water reservoirs are potential sources of deleterious materials downstream, especially during floods or flushing flows (FFs). Their interaction with these events is important for determining potential risks and evaluating management actions. In the Ebro River, the Flix Reservoir accumulated a deposit of more than 3×10^5 t of industrial waste with high Hg concentrations. Because suspended particulate matter (SPM) is the main driver of Hg pollution downriver, this study analyses the transport of particulate matter and Hg pollution from the Flix Reservoir to the Ebro Estuary during FFs. Time series of currents, turbidity and downward particulate matter fluxes were obtained by current meters, turbidimeters and sediment traps assembled in benthic tripods. They were deployed in the reservoir and at two locations in the estuary during two recording periods that each captured a flushing flow (FF) event. In addition, SPM samples were collected during the study period at several locations along the river course, from upstream of the Flix Reservoir down to the river mouth, to measure the suspended particulate matter and associated Hg mobilized downstream.

A continuous background level of Hg pollution was observed during the deployment periods, but the Hg and particulate matter fluxes increased by between one and two orders of magnitude during FFs. Though the two events reached similar water discharges, the first FF was after the wet season and generated lower particulate matter concentrations and fluxes, but higher Hg contents than the second, which occurred after the dry season. The higher available particulate matter in the second event diluted the polluted Hg particle load more than the first event. Thus, similar FFs may result in

different Hg concentration and sediment transport episodes, largely depending on the previous hydrological regime and the river sediment availability. These findings should be considered for FF management.

Keywords: flushing flow, mercury transport, particulate matter, polluted reservoir, salt wedge estuary, Ebro River.

1. Introduction

The uncontrolled dumping of waste and the construction of dams in many rivers around the world have favoured the accumulation of anthropogenic deposits in fluvial systems on a global scale (Wildi et al., 2004; Syvitski et al., 2005; García Bravo et al., 2009; Man et al., 2013; Skalak et al., 2013). With about 800,000 dams around the world, more than 50% of the river sediment flux is trapped in regulated watersheds (Vörösmarty et al., 2003; Nilsson, 2009), and water reservoirs are on the whole an effective global pollution trap (Wildi et al., 2004). These historical deposits are a pollution source that could affect the quality of water used for irrigation and human consumption. The more erodible part of these deposits can leak from the reservoir along with the lighter fine suspended sediment and be transported downstream. In addition, environmental management may induce changes in physicochemical conditions, leading to the release of pollutants (Frémion et al., 2016)

Many pollutants are linked to fine particulate matter ($<63\ \mu\text{m}$) (Feng et al., 1998; Israelsson et al 2014) that can bypass the dams and flow downstream to be

trapped in estuaries due to the convergence of flows associated with their circulation (Yellen et al., 2017; Ralston and Geyer, 2017; Burchard et al., 2018). There, fine particles and pollutants are recirculated and can be periodically deposited and resuspended (Garnier et al., 1991; Grabemann et al., 1997; Turner and Millward, 2002; Geyer and Ralston, 2018).

Under the Mediterranean climate, long periods of low river flow during droughts alternate with flash floods, and most of the river sediment transport takes place during flood events (Batalla et al., 1995; García et al., 2000; Vericat and Batalla, 2006). During the 20th century, many dams were built for water management and hydroelectric energy production, leading to artificial flood management. Controlled flood flows from dams, known as FFs, are used for environmental and/or engineering purposes (Kondolf and Wilcock, 1996; Batalla et al., 2006; Batalla and Vericat, 2009).

In the lower Ebro River (Fig. 1), FFs have been implemented since 2003, mainly to remove the excess of macrophytes, and these events have been monitored, analysed and modelled (Batalla and Vericat, 2009; Tena et al., 2012). FFs have also been suggested as a sediment management tool for increasing the sediment delivered to the coast and delta plain (Rovira and Ibáñez, 2007). During FFs, the SPM concentration doubles that of natural floods, although discharges are typically lower due to their shorter peaks and duration (Tena et al., 2012).

The last reservoir of the Ebro River (the Flix reservoir) is highly polluted by the mercury (Hg) of a waste deposit retained on it since 1948 (Palanques et al., 2014). The effect of FFs on the transport of Hg to the Ebro Estuary has not been addressed up to now. This transport is closely tied to the transport of particulate matter because of the great affinity of Hg for particles (Gunnarson et al., 2009; Masson et al., 2018).

In this paper, we study the effects of two FFs on the particulate matter and Hg transport from the Hg-polluted Flix Reservoir to the Ebro Estuary. Like the Flix Reservoir, many dams in the world were originally built for industrial purposes and now retain polluted sediment (Lenhart, 2003). The interaction of these sediments with river floods and FFs is important for determining their potential risks and evaluating management actions. Moreover, though many studies have characterized contaminants in steady conditions or in sediment records, fewer have considered the transport dynamics of suspended sediment–borne pollution (Quesada et al., 2014; Geyer and Ralston, 2018).

2. Study area and Methods

2.1 Study area

The Ebro River, the second largest river flowing into the NW Mediterranean, drains an area of around 85,000 km² that was progressively impounded during the 20th century and now has close to 190 dams (Vericat and Batalla, 2006). The largest complex of reservoirs in the Ebro River is formed by the Mequinenza (water capacity: 1534 hm³), Riba-roja (water capacity: 207 hm³) and Flix (water capacity: 11 hm³) dams. This complex is located in the lower reaches of the river and impounds 97% of the basin (Fig. 1A). The suspended sediment discharge in the pre-dam period was estimated to be between 18 and 21 x 10⁶ t y⁻¹, whereas in the post-dam period it has only been about 0.1 x 10⁶ t y⁻¹ (Palanques et al., 1990; Guillén and Palanques, 1992; Vericat and Batalla, 2006).

The last downstream dam, that of the Flix Reservoir, is located 110 km upstream from the Ebro River mouth. The Flix dam was built in 1948 only a few hundred metres downstream from an industrial complex located on the south river bank (Fig. 1B). In 1949, this industrial complex was transformed into a chlor-alkali plant using the mercury cathode electrolytic technique, which requires large amounts of mercury and is well-known for its environmental impact (UNEP, United Nations Environment Programme, 2002; OSPAR Commission, 2005; Ullrich et al., 2007). As a consequence, the Flix Reservoir and the lower Ebro River have been affected by the residue dumped from the plant for decades. This residue formed an industrial waste deposit of more than 3×10^5 t that obstructed about 50% of the reservoir water section (Fig. 1C), and Hg is the trace element that shows the highest anomalies, reaching concentrations of up to 640 mg kg⁻¹ (4 orders of magnitude higher than natural levels in river mud) (Palanques et al. 2014). The Hg pollution from the Flix industrial complex affects organisms living in the Flix Reservoir and downstream, such as worms (Ramos et al. 1999), zebra mussels (Carrasco et al. 2008), crayfish (Suarez-Serrano et al. 2010), carps (Navarro et al. 2009) and catfish (Carrasco et al. 2011).

Between the Flix dam and the Ebro Delta plain, the river is gravel-bedded (Vericat and Batalla 2006), and polluted suspended particles flow towards the estuary. There is no significant particulate Hg pollution source downstream from the Flix complex to the estuary. The Ebro River has a microtidal salt wedge estuary, and the salt wedge can intrude more than 30 km when the water discharge decreases below 150-200 m³ s⁻¹. Between this discharge and 350-400 m³ s⁻¹, the head of the salt wedge is generally located at Gracia Island, 15 km from the river mouth, which acts as a topographic threshold for the intrusion. Above about 400 m³ s⁻¹, the salt wedge is pushed out of the river (Guillén and Palanques, 1992).

Dam removal or failure can result in decades of accumulated material being released downstream rapidly and catastrophically (Stanley and Dole 2003). This risk is now being eliminated because the Flix waste deposit has been isolated from the main river course, and decontamination works to remove the waste from the reservoir are underway (Palanques et al., 2014). The data shown in this study were obtained before it was decided to remove the waste deposit from the Flix Reservoir, and the results will be important reference parameters for evaluating the evolution of Hg pollution in the lower Ebro River when the total decontamination of the reservoir has been completed.

2.2. Field work

River particulate matter is retained in the reservoir and the estuary, where it settles. Downward particulate matter was collected by vertical sediment traps to determine its pollutant concentrations and estimate the trapped downward particle fluxes. In addition, as SPM is the main driver of Hg pollution downriver, time series of currents and turbidity data were recorded to assess the transport of particulate matter.

Three instrumented benthic tripods were deployed during two one-month periods in which FFs took place. Deployment 1 was from 12 April to 12 May 2006 and Deployment 2 from 11 November to 11 December 2006. One tripod was deployed in the Flix Reservoir at 3.5 m depth and the other two in the Ebro Estuary near Gracia Island and near the river mouth at 5.50 m depth (Fig. 1A). Each tripod was equipped with an Aanderaa Doppler current meter (RCM-9) with temperature, salinity, 0-25 formazin turbidity unit (FTU) and 0-500 FTU range turbidity sensors, and also with a vertical cylindrical sediment trap (100 cm high and 15.2 cm wide) with 10 receiving

166 cups. The current, temperature, salinity and turbidity sensors were located 60 cm above
167 the bottom, and the top of the trap was located 150 cm above the bottom. The current
168 meter sampling intervals were set at 5 min and the sediment trap collecting interval was
169 set at 3 days for each cup.

170 The sediment trap cups were filled with a borax-buffered 5% formaldehyde
171 solution in 0.20 μm filtered seawater before their deployment. This poisoning solution
172 limits degradation of trapped particles and prevents the mechanical disruption of
173 swimming organisms (“swimmers”) that occasionally enter the traps (e.g., Knap et al.,
174 1996). After recovery, the cups were stored in the dark at 2–4 °C until they could be
175 processed in the laboratory.

176 During the four field campaigns for the two deployments and the two recoveries
177 of the benthic tripods carried out on 11 April, 13 May, 10 November and 12 December,
178 a set of vertical hydrographic profiles were recorded using a Sea-Bird SBE 9 CTD
179 coupled with a Seapoint turbidimeter. Water samples were taken with a Niskin bottle
180 attached to the CTD in surface and near-bottom waters at the tripod sites. Water was
181 also sampled upstream from the Flix Reservoir, near the town of Riba-roja, as a control
182 station to determine the Hg levels of the SPM reaching the Flix Reservoir (Fig. 1A), and
183 along the north and the south banks of the Flix Reservoir (see location in Fig 1C). The
184 November FF was also monitored by taking water samples every 1-2 hours at Riba-roja,
185 at the Flix meander (just after the reservoir), at Mora d’Ebre and at Gracia Island in the
186 estuary (see location in Fig. 1A). Water samples were stored in the dark at 2–4 °C until
187 they could be processed in the laboratory

188 The Confederación Hidrográfica del Ebro provided hourly daily water discharge
189 at the gauging stations of Ascó and Tortosa (see location in Fig. 1A).

2.3 Laboratory work

Settling trapped material was split with a peristaltic dispenser to divide the total sample into several homogeneous aliquots. “Swimmers” were removed from the samples by wet-sieving the sample through a 1 mm nylon mesh. Samples were left to settle again (for one to two days) and examined under a magnifying glass to remove the “swimmer” organisms smaller than 1 mm. By this method, the bulk of swimmers were removed and the “biological” contamination, if any, was negligible (Heussner et al., 2006). Subsequently, the samples were washed with Milli-Q water and centrifuged three times to extract all the remaining seawater. Finally, the samples were freeze-dried by lyophilisation and weighed. They were then homogenized in an agate mortar for the analysis of Hg. Downward total mass flux (TMF) was computed using the total mass weight divided by the trap collecting area (182 cm^2) and the sampling interval (3 days).

Water samples were filtered onto Whatmann glass microfibre filters (GFFs) to obtain the SPM concentration and suspended sediment samples for analysis of their Hg content. Before use, the GFFs were rinsed with Milli-Q water, placed for 24 hours in an oven at 550° Celsius, allowed to cool for another 24 h, and then weighed. After filtering of the water samples, the filters with the suspended sediment samples were dried to constant weight at 40° Celsius and then placed in a desiccation bowl for 24 h. The SPM concentration was estimated by dividing the weight of the suspended sediment by the volume of filtered water in each sample.

The turbidity data from the tripods and the CTDs were recorded in FTU and converted into estimates of SPM concentration through regression curves from paired measurements of turbidity and SPM concentration obtained with water samples taken with the Niskin bottle coupled with the CTDs at the tripod sites. In the estuary the calibration curve was

$$\text{SPM concentration} = 0.94 \text{ FTU} - 1.99 \text{ (R}^2 = 0.66\text{)}$$

and in the reservoir it was

$$\text{SPM concentration} = 2.05 \text{ FTU} - 12.32 \text{ (R}^2 = 0.57\text{)}.$$

As the regression curves were not highly significant and turbidity-SPM concentration rating curves can vary in time, reported values from the time series should be considered estimates of SPM concentration in order to visualize the temporal evolution of this parameter throughout the deployment period.

The Hg content of the downward and suspended particulate matter samples was analysed using a LECO AMA254 Mercury Analyzer complying with the US EPA Method 7473 (US EPA, 2007). The instrument's detection limit of Hg is 0.01 ng. PACS-2 certified reference material from the National Research Council Canada and blank samples were used for analytical quality control. Thirty-five replicate determinations of the certified reference material with a certified value of $3.04 \pm 2.0 \text{ mg kg}^{-1}$ (mean and SD) gave an average value of $2.98 \pm 0.1 \text{ mg kg}^{-1}$. Downward Hg fluxes were obtained by multiplying downward TMF by the Hg concentrations of the trapped particle samples.

3. Results

3.1. River water discharge

River water discharge during the two FFs reached similar maximum values: 1498 m³ s⁻¹ in May and 1539 m³ s⁻¹ in November at the Ascó gauging station and 968 m³ s⁻¹ in May and 895 m³ s⁻¹ in November at the Tortosa gauging station. The lower peak at the Tortosa station was a consequence of the change in the shape of the hydrograph due to the floodwave attenuation downstream. The duration of these events was also similar, in both cases about 15 h. In the rest of the deployment periods, the river discharge ranged between 120 and 200 m³ s⁻¹ (Fig. 2).

In this paper, because the Ascó gauging station is closer to the Flix Reservoir and the Mora d'Ebre sampling sites and Tortosa is the closest gauging station to the estuary (Fig. 1A), we will refer to the Ascó water discharge for the Flix and Mora d'Ebre data and to the Tortosa water discharge for the estuary data.

The context in which the two FFs occurred was different. The first FF took place one month after a period of high water discharge (>400 m³ s⁻¹) and a natural flood (up to 1530 m³ s⁻¹), whereas the second FF took place after six months of low water discharge (mainly <200 m³ s⁻¹) (Fig. 2).

3.2. Hg concentration in SPM along the lower Ebro River

Hg concentrations in the SPM upstream of the Flix Reservoir at the Riba-roja station ranged between 0.07 and 0.43 mg kg⁻¹, with a mean value of 0.19 mg kg⁻¹ (Table 1; Fig. 3). These are the Hg levels of the SPM reaching the Flix Reservoir, which are relatively low. At the south bank of the Flix Reservoir, over the waste deposit, the Hg levels of the SPM were between two and four orders of magnitude higher than those at Riba-roja (Table 1; Fig. 3). On the north bank, however, where the riverbed is not covered by the industrial waste, the Hg levels were only one order of magnitude higher than upstream of the Flix Reservoir (Fig. 3; Table 1), suggesting that the particles leaving the waste deposit probably reach the deeper channelized part of the reservoir (Figs. 1C) and are mixed with the upstream inputs, diluting the pollution concentration.

Downstream of the Flix dam, Hg concentrations were similar to those on the north bank of the reservoir (Fig.3; Table 1). In the estuary, Hg values decreased slightly, but still maintained Hg concentrations about one order of magnitude higher than upstream of the reservoir (Fig. 3; Table 1), possibly indicating that the polluted particles escaping from the waste deposit and mixed with the upstream inputs bypass the dam and are transferred directly downstream to the estuary.

In the estuary, an upper fresh water layer up to 2 m thick and a sharp horizontal gradient to the salt water below were observed during the four field campaigns to install and recover the tripods. At Gracia Island, the SPM and Hg concentrations tended to be slightly higher in the surface fresh water than in the bottom salt water, whereas at the river mouth, surface and near-bottom values were more similar (Supplementary Material Fig. 1).

During the November 21 FF, Hg concentrations of SPM upstream of the reservoir maintained relatively constant values (around 0.16 mg kg⁻¹). However,

downstream of the reservoir the concentrations increased to 2.40 mg kg^{-1} at the Flix meander, to 2.25 mg kg^{-1} at Mora d'Ebre and to 0.80 mg kg^{-1} at Gracia Island (Fig. 4). These increases lasted only two hours and took place at the beginning of the FF, when more erodible polluted sediment was available for the increasing currents, and before the SPM concentration and water discharge peaks. The Hg concentration decreased when the SPM concentration increased at the peak of the flow, when most sediment removal from bed and bank erosion occurred along the river, thus diluting the pollution load.

Discharge of SPM-borne pollution can be estimated where gauging stations measure the volumetric flow rate of water that is transported through a given river cross-sectional area, assuming homogeneous pollutant and SPM concentrations across it. In the case of the November FF event, Hg discharge can be roughly estimated extrapolating the water discharge from the Ascó gauging station and from the SPM and Hg concentrations monitored at Riba-roja and Mora d'Ebre during this event (Supplementary Material Fig. 2). The maximum Hg discharge was 18 mg s^{-1} at Riba-roja (upstream of the reservoir), and 672 mg s^{-1} at Mora d'Ebre (downstream of the reservoir).

At Gracia Island, the salinity record indicates that the fresh river water occupied the whole estuary section during the November FF event, so the Hg discharge was roughly estimated in the same way, but the water discharge was extrapolated from the Tortosa gauging station, giving a maximum Hg discharge of 156 mg s^{-1} (Supplementary Material Fig. 2).

From the SPM samples taken when the river discharge was between 120 and $200 \text{ m}^3 \text{ s}^{-1}$, we can also estimate Hg discharges ranging from 0.2 to 0.4 mg s^{-1} at Riba-roja

and from 0.8 to 2.4 mg s⁻¹ at Mora d'Ebre. All these data confirm the downriver transport of Hg from the reservoir, especially during FFs.

3.3. Time series of hydrodynamics, SPM concentration and particle matter fluxes

3.3.1. The Flix Reservoir

In the Flix Reservoir, daily oscillations of current speed (from 1 to 15 cm s⁻¹) and SPM concentrations (from 1 to 30 mg L⁻¹) were recorded during most of the deployment periods, and were probably induced by the dam's water management (Fig. 5). However, current speed and SPM concentrations peaked to 23 cm s⁻¹ and 411 mg L⁻¹ in the May FF and to 37 cm s⁻¹ and 980 mg L⁻¹ in the November FF, respectively. Downward TMF was between 6 and 27 g m⁻² d⁻¹ during most of deployment 1 and between 50 and 100 g m⁻² d⁻¹ during most of deployment 2, increasing to 105 g m⁻² d⁻¹ in the sample affected by the May FF and to 390 g m⁻² d⁻¹ in the sample affected by the November FF (Fig. 5). Therefore, SPM concentration and downward TMF were two to four times higher during the November FF than during the May FF. Recorded peaks of SPM concentrations and downward particle fluxes corresponded mostly to polluted sediment resuspended at the reservoir during the FFs.

3.3.2. The Ebro Estuary

During most of the two deployment periods, time series at the estuary stations were recorded in the salt wedge intruding the estuary, where near-bottom currents ranged between 1 and 15 cm s⁻¹, alternating from up-estuary (~270°) to down-estuary

(~90°) (Figs. 6 and 7). Under these conditions, the SPM concentration ranged between 1 and 12 mg L⁻¹ and downward TMF between 5 and 40 g m⁻² d⁻¹. However, during the May and November FFs, current speed at Gracia Island increased to over 50 cm s⁻¹, flowing seawards and pushing the salt water and the sediment retained in the estuary downriver for 1-2 days (Fig. 6). At this site, SPM concentration and downward TMF increased by between one and two orders of magnitude during the FFs, and these values were around twice as high in the November FF as in the May FF (Fig. 6, and Table 2).

At the river mouth, the FFs induced lower peaks of near-bottom currents (32 and 38 cm s⁻¹) and short periods (only up to six hours) of near-bottom downriver flow of brackish waters (salinity of 24.06 in spring and 9.52 in autumn) (Fig. 7). The near-bottom SPM concentration and downward TMF did not increase during the May FF, and the SPM concentration increased very sharply to 184 mg L⁻¹ during the November FF, but just for two hours.

3.4. Time series of Hg concentrations and fluxes of the trap-collected particulate matter

Hg concentrations of the downward particulate matter at the Flix Reservoir site were between two and three orders of magnitude higher than natural values and were higher in the first deployment than in the second (Fig. 8). The FFs resuspended polluted reservoir sediment and increased the downward Hg fluxes about four times, reaching 2020 and 4860 µg m⁻² d⁻¹ in the sampling periods coinciding with the May and November FFs, respectively (Fig. 8).

In the Ebro Estuary, the downward Hg concentration during the two deployment periods were almost one order of magnitude higher than natural values and were higher

in the first deployment than in the second. (Fig. 8). At Gracia Island, the FF events increased the downward Hg fluxes by more than one order of magnitude in the corresponding trap samples, reaching a similar value in both FFs (Fig. 2; Table 2). By contrast, there were no increases in the downward Hg fluxes linked to the FFs at the river mouth (Fig. 8).

The sediment trap samples showed no clear increase in the Hg concentration induced by the FFs (Fig. 8). The two-hour increase detected during the November FF monitoring (Fig. 4) was probably masked in the sediment trap samples because they include three-day periods of downward particles.

4. Discussion

4.1 Comparison of the May and November FFs

Sediment dynamics in regulated rivers is a function not only of the hydrology during the events, but also of several parameters arising from the preceding conditions, such as sediment availability, exhaustion and flashiness, and loss of energy due to flow routing (Tena et al., 2012). In the Ebro River, the May and November FFs sharply increased current speed, SPM concentration and downward particle and Hg fluxes (Figs. 5, 6, 7, 8). These events were similar in water discharge and duration, but the SPM concentration and downward TMF were lower and the Hg concentration higher during the May FF than during the November FF (Figs 5, 6, 7, 8; Supplementary Material Fig. 3; Table 2). Batalla and Vericat (2009) measured a maximum SPM concentration of

mg L⁻¹ during the November FF and of only 88 mg L⁻¹ during the May FF, with mean values of 244 and 56.7 mg L⁻¹, respectively, thus corroborating the lower sediment transport during the May FF.

The different sediment transport during the two FFs could be explained by the river conditions before each event (Fig. 2). The May FF occurred after the wet season. The previous natural floods could have transported the more erodible sediment downstream, winnowing the river and the reservoir, pushing back the salt wedge and leaving a lower fine sediment load available to be transported along the river and estuary during the first deployment and FF. This reduction of the sediment availability probably decreased the dilution of the Flix polluted particles with upstream particles, giving higher Hg concentrations in May than in November (Fig. 8, Tables 2 and 3).

By contrast, the November FF was in the dry season after six months of low water discharge ($< 200 \text{ m}^3 \text{ s}^{-1}$), which allowed a higher sediment load to be stranded in the river, in the reservoir and in the estuary, where the salt wedge intrusion is maintained at discharges below about $400 \text{ m}^3 \text{ s}^{-1}$ (Guillén and Palanques 1992). All this would have left more sediment available to be transported during the November FF than during the May FF. The higher availability of low-polluted particles from upstream of the Flix Reservoir in November may have diluted and decreased the Hg concentrations of the trap-collected particulate matter more than in May (Fig. 8; Tables 2 and 3). One fact that supports this hypothesis is that downward TMF in the May FF at Gracia Island was approximately half that in the November FF (Fig. 6; Table 2), but Hg concentrations in the May event were almost double those in the November event, so the resultant downward Hg flux at Gracia Island was similar during the two events (Fig. 8, Table 2).

4.2 Effects of FF in the particulate matter and Hg transport of the estuary

In estuaries, currents and SPM concentrations can change strongly with depth and also across and along them according to the interactions between marine and freshwater. Therefore, the estimate of the total SPM and Hg discharges requires data about the vertical and lateral structure of the velocity and SPM distributions, which are difficult to obtain. However, an approach can be made by estimating the integrated sediment transport calculated from the time series of current velocity and SPM concentrations at a single location, which provides an indication of the temporal variability of sediment transport at a single point (Geyer and Ralston, 2018). Assuming that particles move with the velocity of the water within which they are suspended (Wright, 1995), the instantaneous near-bottom sediment flux (q) in $\text{g m}^{-2} \text{s}^{-1}$ at the height of the instrument is obtained as the product of the velocity module c and the SSC, in mg/L :

$$q(t) = c(t)SSC(t)$$

The integration of the instantaneous horizontal fluxes for the duration of the deployments yielded the cumulative horizontal suspended sediment transport in kg m^{-2} .

The cumulative horizontal SPM transport at the estuary site of Gracia Island indicates that there was a dominant up-river inflow of SPM in the salty layer during the salt wedge intrusion periods (104 and 293 kg m^{-2} during D1 and D2 respectively; Supplementary Material Fig. 3). This helped retain fine sediment and associated Hg inside the estuary during low water discharge periods, leaving them available to be transported during FFs and natural floods. By contrast, the cumulative horizontal SPM

transport during the FFs was down-estuary and increased by between one and two orders of magnitude, and twice more in the November event than in the May event (3764 and 9535 kg m⁻² respectively; Supplementary Material Fig. 3 and Table 2).

Another rough estimation of the sediment transport in the estuary can be made from the SPM concentration recorded by the Gracia Island turbidimeter and the water discharge from the Tortosa gauging station during the FFs, when the river water occupied the whole estuary section. The SPM discharges estimated in this way show a maximum value in the November FF (352 kg s⁻¹) almost twice that in the May FF (190 kg s⁻¹).

An approach to estimate the cumulative suspended Hg transport can be made from the cumulative SPM transport with the assumption that the Hg concentration of the trap samples collected during the FFs are the mean Hg concentration of the SPM during these events. These calculations give a Hg transport increase of two orders of magnitude during both FFs (from about 0.06 g m⁻² to a maximum of 3.61 g m⁻² in the May FF and 3.99 g m⁻² in the November FF).

Following the same assumption for the mean Hg concentrations during the FFs, another approach can be made to estimate the Hg discharge during these events, considering the SPM concentration recorded by Gracia Island turbidimeter and the water discharge from the Tortosa gauging station. These calculations give similar maximum Hg discharges of 173 g s⁻¹ in the May FF and 166 g s⁻¹ in the November FFs.

4.3. Implications of the FFs in the estuary

Although these SPM and Hg transport estimates have uncertainties, they suggest that similar maximum suspended Hg fluxes and discharges were reached during both FF's, as the downward Hg fluxes also did (Fig. 8). However, the higher sediment load transported during the November event diluted more the Hg concentration of the particulate matter transported to the estuary than the May FF. This has implications for the assessment of toxicity risks. Long et al. (1995) defined two values for Hg to establish three ranges according to which the effects to the environment are predicted to be minimal (value $< 1.5 \text{ mg kg}^{-1}$), occasional (1.5 mg kg^{-1} value $< 0.71 \text{ mg kg}^{-1}$) or frequent (value $> 0.71 \text{ mg kg}^{-1}$). In November, Hg concentrations were within the range of occasional effects but in May they were within the range of frequent effects. This kind of effects should be considered for FF management.

The absence of major suspended and downward particle flux increases at the river mouth station during the FFs (Fig. 7; Supplementary Material Fig. 3; Table XX) suggests that the sediment outflow at this site was mainly through the upper freshwater layer. This phenomenon may be favoured by the downstream thinning of the freshwater layer and the higher flow speed in this layer (Guillén and Palanques 1992), which prevents the mixing of particles between fresh and salt layers.

FFs eject downstream the salt water at the head of the estuary (near Gracia Island), but in the two cases studied they did not last long enough to push out the salt water from the river mouth. This could also mean that part of the sediment transported by the FFs did not reach the river mouth, remaining in the estuary until new, larger floods transferred it to the marine environment. This is important for managing river sediment transport, because it could indicate that stronger or longer flows must be generated if the purpose of the FF is not only to remove the excess of macrophytes but also to transfer river sediment into the coastal environment.

Hg discharges from chlor-alkali plants have also spread into other river and estuarine systems such as the Alveiro Lagoon, the Mersey Estuary and the Penobbscot Estuary. Hg concentrations in suspended sediment of the Alveiro Lagoon ranged between 0.46 and 6.7 mg kg⁻¹ at the mouth of the channel discharging from the plant and between 0.33 and 1.7 mg kg⁻¹ about 1-2 km down-estuary (Pereira et al., 1998). In the Mersey Estuary, the Hg concentration of the surface bottom sediment was between 0.6 and 1.0 mg kg⁻¹ (Harland et al., 2000) and in the Penobscot Estuary it was between 0.12 and 2.75 mg kg⁻¹ (Merritt and Amirbahman, 2007; Yeager et al., 2018). The Ebro estuary has a lower tidal range than the above estuaries and, unlike them, it has a dam downstream from the pollution source.

Increases in sediment transport and pollutant load during FFs and natural floods have also been recorded in the Soča River in the Gulf of Trieste. This river was affected by the Hg pollution from a former mine in Idrija, and a flood of similar characteristics to the studied ones (1600 m³ s⁻¹) increased the Hg concentration of SPM to 46.6 mg kg⁻¹ (Rajar et al., 2000). This is one order of magnitude higher than downriver from the Flix reservoir, probably because, independently of the pollution sources, the Soča River had no dam downstream of the mine. The Flix dam, which was built just downriver of the chlor-alkali plant, retained the industrial waste in the reservoir and limited the release of polluted particles downstream. This dam reduced the transport of the polluted load towards the estuary, discharging it episodically or leaking it in continuous lower fluxes.

The Hg concentrations detected in the Ebro Estuary were similar to those detected in the Rhone Delta during the 1980s, ranging mainly from 0.6 to 0.9 mg kg⁻¹ (Figueres et al., 1985, Cossa and Martin, 1991), but they were significantly higher than those detected at the Beaucaire gauging station (located 60 km upstream from the Rhone River mouth) between 2011 and 2016, which showed a mean Hg concentration

of 0.095 mg kg⁻¹ (Poulier, 2019). This means that, over the past 30 years, major improvements in wastewater treatment through implementation of the Urban Wastewater Treatment Directive (UWWTD) have helped reduce the pollutants in the Rhone River (Olivier et al., 2008), whereas in the Ebro River, the industrial waste deposit of the Flix Reservoir is a historic Hg source that still needs to be removed.

A significant decrease in the Hg pollution in the Ebro Estuary is expected after the decontamination of the Flix Reservoir. However, in some estuaries the bidirectional transport of sediments and the estuarine regime leads to a long dilution time scale, as occurred in the Penobscot Estuary, where mercury released into the river in the 1970s now has a nearly uniform concentration in mobile SPM and a residence time of approximately 25 years. (Geyer & Ralston 2018).

5. Conclusions

This study indicates that polluted sediments accumulated in reservoirs can become a permanent source of pollutants downstream and can either be discharged episodically or leak continuously in lower fluxes towards estuarine systems, where they are recirculated.

The FFs in the lower Ebro River sharply increased the SPM concentration, the particulate matter fluxes and the Hg fluxes in the reservoir and in the estuary at Gracia Island. The Hg concentration also increased but only during the first 2 hours of the November FF. Most of the downriver particulate matter and Hg transport in the estuary at Gracia Island occurred during the FFs.

Similar FF events can produce different effects in SPM and particulate Hg transport, depending on the previous dynamics of the river system. After a long low water discharge period, the November FF generated higher particulate matter fluxes than after a high river discharge period in the May FF, mainly due to higher sediment availability in the November event. However, the Hg concentration was lower during the November FF because the higher available particulate matter diluted the polluted Hg particle load more than during the May event and the resultant Hg fluxes in both events were similar.

The reduction of the Hg concentration when there is more particulate matter availability in the river to dilute the pollution load should be considered for FF management. In addition, this study gives reference parameters for controlling the evolution of Hg pollution in the lower Ebro River after the decontamination of the reservoir, in order to determine how the system responds to the elimination of this pollution source.

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	Riba-roja	Reservoir South	Reservoir North	Mora D'Ebre	Estuary Gracia Island	Estuary River Mouth
Max	0.43	173.69	3.64	3.99	3.13	1.41
Min	0.07	7.24	0.75	0.83	0.62	0.66
Mean	0.19	41.62	1.61	1.93	1.01	0.85
Sd Dv	0.11	10.39	0,89	0.53	0.53	0.02

749

750 Table 1. Maximum, minimum, mean and standard deviation of the Hg concentrations
751 (in mg kg⁻¹) in suspended particulate matter samples from Riba-Roja, from the Flix
752 Reservoir on the south bank (Reservoir South) where the industrial waste deposit was
753 accumulated (RS in Fig. 1C), from the Flix Reservoir on the north bank (Reservoir
754 North) opposite the industrial complex (RN in Fig. 1C), from Mora d'Ebre, from Gracia
755 Island and from the river mouth (Location in Fig. 1A).

756

	Gracia Island		River Mouth	
	May FF	November FF	May FF	November FF
D. TMF (gr m ⁻² d ⁻¹)	286.0	569.5	11.1	91.1
Max. SPM conc. (mg L ⁻¹)	298.0	557.5	6.7	184.7
Cum. SPM Flux (T m ⁻²)	3.74	8.8	0.05	0.08
D. Hg conc. (mg kg ⁻¹)	0.86	0.47	0.71	0.55
D. Hg Flux (µg m ⁻² d ⁻¹)	246.8	268.9	7.9	50.6

757

758 Table 2: Downward total mass flux (D. TMF), maximum SPM concentration (Max.

759 SPM conc.), cumulative suspended particulate matter flux (Cum. SPM Flux), downward

760 Hg concentration (D. Hg conc.) and downward Hg flux (D. Hg Flux) during the

761 flushing flow sampling period.

Figure 1

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Figure 1. A) Google Earth image of the lower Ebro River showing the locations where the benthic tripods were deployed (yellow stars), where the water samples were taken (orange triangles and yellow stars), and where the two gauging stations of the Ebro Water Authorities (Confederación Hidrográfica del Ebro) were installed (Ascó and Tortosa) (green circles). B) Google Earth image of the Flix Reservoir and the Flix meander before the extraction of the waste deposit. C) Bathymetry of the Flix Reservoir before the extraction of the waste deposit. RS: Reservoir South bank, where the industrial waste deposit (pale orange surface) has accumulated. RN: Reservoir North bank. HPP. Channels: underground hydraulic power plant channels. Orange triangles: water sampling stations. Yellow star: tripod position. Coordinates are in UTM, zone 31.

Figure 2
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Figure 2: Ebro River water discharge from February to December at the Tortosa and Ascó gauging stations. D1: Deployment 1, from April 12 to May 12. D2: Deployment 2, from November 11 to December 11. Zooms correspond to the May FF and the November FF.

Figure 3: Hg concentrations in the SPM sampled along the lower Ebro River at the Riba-roja (R.roja), the Flix Reservoir (Flix R), Mora d'Ebre (M. d'Ebre), Gracia Island (Gracia I.) and river mouth (R. Mouth) stations. White rhombuses at the Flix Reservoir correspond to Hg values in the SPM from the north bank, opposite the industrial complex (RN in Fig 1C and Reservoir North in Table 1), whereas black points are from the south bank, where the industrial waste deposit was accumulated (RS in Fig 1C and Reservoir South in Table 1). Mean values (red crosses). Standard deviation (green lines).

Figure 4
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Figure 4. Time series of water discharge at the Ascó and Tortosa gauging stations along with the Hg concentration and SPM concentration (SPM C) measured at Riba-roja, the Flix meander, Mora d'Ebre and Gracia Island during the November FF.

Figure 5
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Figure 5: Time series of water discharge at the Ascó gauging station along with temperature, current direction, current speed, SPM concentration (SPM conc.) and downward total mass flux recorded in the Flix Reservoir during deployments 1 and 2. Note the two different scales of SPM concentration. D1: deployment 1. D2: deployment 2. FF: flushing flow

Figure 6

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Figure 6: Time series of water discharge at the Tortosa gauging station along with salinity, current direction, current speed, SPM concentration (SPM conc.) and downward total mass flux recorded at the Gracia Island station during deployments 1 and 2. Note the two different scales of SPM concentration. D1: deployment 1. D2: deployment 2. FF: flushing flow.

Figure 7

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Figure 7: Time series of water discharge at the Tortosa gauging station, salinity, current direction, current speed, SPM concentration (SPM conc.) and downward total mass flux recorded at the river mouth station during deployments 1 and 2. Note the two different scales of SPM concentration. D1: deployment 1. D2: deployment 2. FF: flushing flow

Figure 8
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Figure 8: Time series of Hg concentration and Hg fluxes in the trap-collected particles of the Flix Reservoir, Gracia Island and the river mouth during the two deployment periods. FF: flushing flow.

Supplementary Material Figure. 1

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Supplementary Material Figure 2

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