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## Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France

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Marina Coquery

Abstract: The Rhône River (France) has been used for energy production for decades and 21 dams have been built. To avoid problems due to sediment storage, dam flushing operations are periodically organized. The impacts of such operations on suspended particulate matter (SPM) dynamics (resuspension and fluxes) and quality (physico-chemical characteristics and contamination), were investigated during a flushing operation performed in June 2012 on 3 major dams from the Upper Rhône River. The concentrations of major hydrophobic organic contaminants (polychlorinated biphenyls, polycyclic aromatic hydrocarbons - PAHs, bis(2-ethylhexyl)phthalate [DEHP] and 4-n-nonylphenol), trace metal elements, particulate organic carbon (POC) and particle size distribution were measured on SPM samples collected during this event as well as on those obtained from 2011 to 2016 on a permanent monitoring station (150 km downstream). This allows to compare the SPM and contaminant concentrations and fluxes during the 2012 dam flushing operations with those during flood events and baseflow regime. At equal water discharge, mean SPM concentrations during flushing were on average 6-8 times higher than during flood events recorded from 2011 to 2016. While of short duration (19 days), the flushing operations led to the resuspension of SPM and contributed to a third of the mean annual SPM flux. The SPM contamination was generally lower during flushing than during baseflow or flood, probably due to the fact that flushing transport SPM only issued from resuspended sediment, with no autochthonous particles nor eroded soil. The only exception are PAHs and DEHP with higher concentrations during flushing, which must be issued from the resuspension of legacy-contaminated sediments stored behind the dams before the implementation of emission regulation. During flushing, the variations of POC and contaminant concentrations are also mostly driven by particle size. Finally, we propose a list of recommendations for the design of an adequate monitoring network to evaluate the impact of dam flushing operations on large river systems.



# Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France

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Cadarache, 8 November 2019

Dear Editor,

Please find enclosed the revised version of the manuscript entitled “Impact of dam flushing operations on sediment dynamics and quality in the upper Rhône River, France” by H. Lepage and co-authors.

This manuscript was corrected according to the reviewers recommendations. Answers to their remarks were compiled in a document enclosed with the manuscript. Finally, the total length of the article was also reduced by 10% (from 6752 words to 6078 words excluding references).

Kindly acknowledge the receipt of the same.

Yours sincerely,

On behalf of the authors,

**(H. LEPAGE)**

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\*Response to Reviewers

Comment from reviewers	Answer from corresponding author	Correction in the manuscript
<b>Reviewer 1</b>		
Highlights	corrected	Sediment dynamic differed during flushing operations and flood events
Line 15	corrected	The Rhône River (France) has been used for energy production for decades and 21 dams have been built.
Line 29 – In general it is not advisable to start a sentence with an acronym, rather right it out in full	corrected	The SPM contamination was generally lower than during baseflow and flood regimes.
Line 131	corrected	More than $3 \times 10^6 \text{ m}^3$ of SPM were thus stored in the Verbois reservoir between 2003 and 2012 (Services industriels de Genève, 2014).
Line 133	corrected	[...] which transports $\sim 0.7 \times 10^6 \text{ m}^3$ of SPM per year (approx. 1 to 3 tons dry weight) [...]
Line 309-314 – This section reads as introduction or as part of the discussion	According to reviewer 4, this section was moved in the Materials and Method part.	<b>2.7. SPM and contaminant fluxes calculation</b> One important question regarding the impact of flushing operations is to determine their relative contribution to SPM transport compared to flood events. Such a comparison requires an estimation of the influence of both types of events over a longer time scale. The Jons station allows an estimation of the annual SPM fluxes from 2011 to 2016 (based on hydrological years, i.e. from September to August).
<b>Reviewer 4</b>		
[...] the manuscript need a deep re-organization in order to better address the main aim. In particular, both methodological and research aspects are mixed in the text, and this creates some confusion in the reader. For example, to my opinion comparison between results obtained	According to reviewer 1, the paper is well written and logically organized. However, the comparison between CFC and PT was indeed confusing and a part of this was moved to the supplementary materials (Supplementary Material #1.1)	

<p>with CFC and PT sampling methods is redundant and may be moved to Supplementary material. As well, the final recommendations for an adequate monitoring network of flushing operations doesn't add any relevant information to the previous text.</p>	<p>Regarding the recommendations, we thought that it is interesting to share our feedback on flushing operations monitoring as we did not find any similar study in the literature with a long term monitoring. We also noticed that recommendations are used in scientific papers, even in Journal of Environmental Management (ex :</p> <p><a href="https://doi.org/10.1016/j.jenvman.2019.06.092">https://doi.org/10.1016/j.jenvman.2019.06.092</a>  <a href="https://doi.org/10.1016/j.jenvman.2015.08.014">https://doi.org/10.1016/j.jenvman.2015.08.014</a>  <a href="https://doi.org/10.1016/j.jenvman.2019.109405">https://doi.org/10.1016/j.jenvman.2019.109405</a></p> <p>Since the reviewer 1 did not complain on that point and since specific areas of interest of JEM includes: “Development of methods for environmental quality management (new procedures, characterization techniques, <b>monitoring methods</b>)”, we really think that it is relevant to keep such recommendations in the paper.</p>	
<p>By contrary, some scientific aspects should be more deepened and explained. In the present form, results are mainly presented as a report regarding this particular case-study. The case-study of the Rhone River has been widely studied before in many aspects, as proved by the publications reported in the reference section (e.g. Peter et al. 2014 regards the same flushing event, with samples collected in different sampling stations upstream Jons). Thus, results may be shortened, while in the discussion section a wider generalization and comparison with other</p>	<p>The Rhône River has been indeed studied for decades by many research fields, including sedimentary aspect. However, studies investigating the impact of flushing operations on SPM quality remain rare contrary to what is said by reviewer 4. Furthermore, studies on particulate contaminant behaviors during flushing operations are also very rare at a worldwide scale, as described in the introduction of our paper. Reviewer 4 indicates that Peter et al. 2014 worked on the same event. This is right (and we cite this work in our manuscript) but their approach was</p>	

<p>published cases should be added.</p>	<p>really different with a specific focus on the impact of the flushing operations on benthic invertebrates. Also, their investigation on the water quality was complementary to our study as they conducted measurements of dissolved and particulate metals (Al, Co, Cr, Cu, Fe, Mn, Ni, Pb) while we measured particulate organic contaminants (PCBs, PAHs, DEHP and 4-n-nonylphenols) in addition to particulate metals (Cd, Cu, Pb, Zn, Hg). Finally, the impact of the flushing operations on SPM and associated contaminant over a long term monitoring was not studied by Peter et al. 2014. Therefore, we think that both studies are complementary.</p>	
<p>Moreover, a better explanation of the different behavior of contaminants may be addressed.</p>	<p>Additional information were added to the manuscript according to the comments below (chapter 4.2 and 4.3).</p>	
<p>Abstract: methods are missing. Line 29: what do you mean with "origin"? Lines 30-31 seem to contradict lines 29. Maybe this part should be re-written.</p>	<p>The abstract was entirely rewritten to fit these comments.</p>	
<p>the English text should be revised (e.g. highlights contain some mistakes)</p>	<p>The manuscript was proof-corrected by a native English (certificate enclosed in the built pdf) and Reviewer 1 said that the paper was well written. However, special attention was given in re-writing the manuscript.</p>	
<p>line 84: "fixed" doesn't seem correct: maybe "adsorbed" or "carried by"</p>	<p>This sentence was mixed with another to shorten the manuscript.</p>	<p>Additionally, we still need a better understanding of contaminants remobilization and transport processes under such conditions (Hauer et al., 2018; SedNet, 2014), as floods and dam flushing operations are major events able to transport a large fraction of contaminant fluxes (Poulier et al.,</p>



		2019).
In Figure 1 many other smaller dams are present between Genissiat dam and the sampling station at Jons: do they have some influence on your results (e.g. flux calculations or contaminant concentrations?) Are there contaminated sediments stored in those smaller reservoirs which may be remobilized during the flushing events?	The text was corrected in order to include the specific management of these dams. Unfortunately we have no data on contamination levels in these smaller reservoirs.	Five small dams located between Génissiat and Jons (Figure 1) were also opened during this period and managed in order to prevent both sediment deposition and resuspension. The by-passed sections of the Rhône River (Old Rhône reaches) were disconnected and the whole SPM flux transiting through the reservoirs and tailrace canals. The levels of the reservoirs were lowered as much as possible to prevent deposition and ensure a quick transfer of water and SPM.
lines 100-114: this part seems a summary of the research: the final aim of the research should be more addressed	Additional information was added to clarify the aim of the research.	This study aimed at characterizing the impact of flushing operations on SPM dynamics and quality in a large river system: the Rhône River basin. Thanks to a specific monitoring, the fluxes and mass balances of SPM and associated contaminants triggered by flushing operations in 2012 and 2016 were estimated at different time scales. The variations in contaminant concentrations were related to the characteristics of SPM (particle size distribution, organic carbon content) as well as to their origin in the watershed (eroded soil versus resuspended sediment).
line 119: 95600 km <sup>2</sup> is the watershed area?	Corrected.	The Rhône River (95 600 km <sup>2</sup> watershed area, mean water discharge of $\sim 1700 \text{ m}^3 \cdot \text{s}^{-1}$ at the outlet station of Beaucaire)
lines 152-153: add "2016" in the dates	Corrected	(from May 20 <sup>th</sup> , 2016 at 12:00 to May 31 <sup>st</sup> , 2016 at 12:00).
lines 155-158: some results are based on water discharge values: maybe a short description of this hydrodynamical model performance/validation should be mentioned	Additional information was added	There is no hydrometric station close to Jons and hourly water discharge was calculated using the 1-D hydrodynamical model MAGE (Irstea, France) and discharge inputs from upstream hydrometric stations (Lagnieu), Bourbre (Tignieu-Jameyzieu),

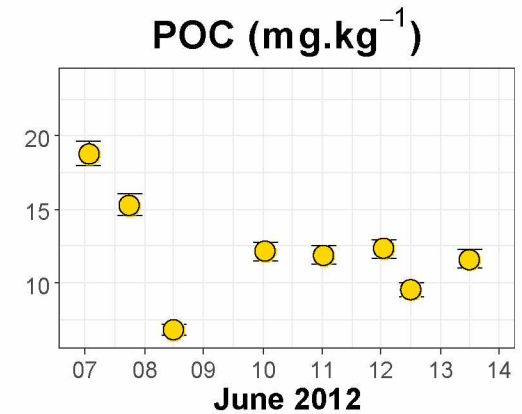
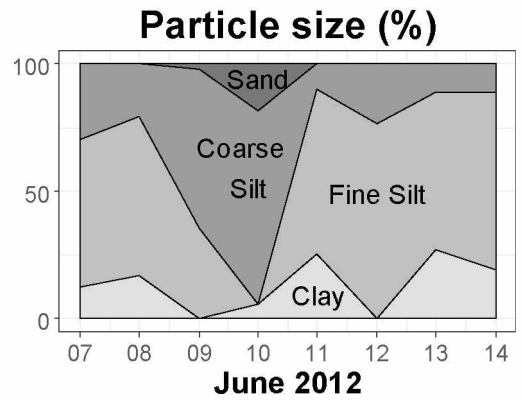
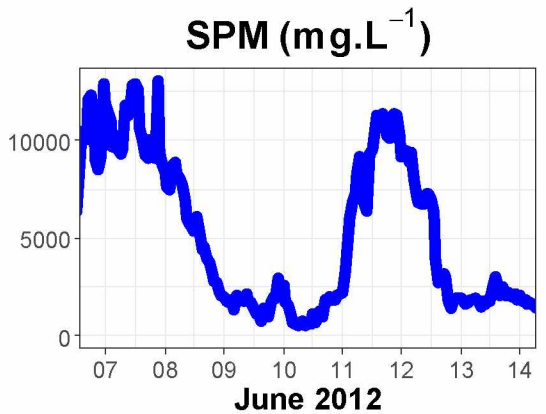
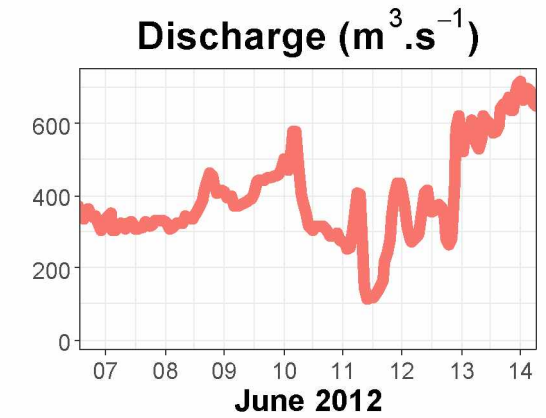
		and Ain (Port-Galland) Rivers (Figure 1) (Dugué et al., 2015; Launay et al., 2015). This model is calibrated for the whole Rhône River and outputs were evaluated against measured water levels, with a maximal accepted difference of 10 cm.
lines 205-206 should be moved to discussion or supplementary material	Part 2.4 including lines 205-206 was moved to supplementary materials to reduce manuscript length.	
- A "Data analysis" paragraph is missing. Maybe lines 309-314 could be part of this paragraph. As well, the calculation of fluxes should be reported	Lines 309-314 were moved to the Materials and Method part (chapter 2.7). Flux calculation was already explained in this chapter.	
lines 292-308 are partly a repetition of paragraph 3.3, so this part can be moved to paragraph 3.3 and shortened	Lines 292 to 308 describe the variation of the SPM parameters (POC, particles size distribution) and contaminants concentrations <u>during the flushing operations</u> , while part 3.3 describe the variation <u>between the different hydrological conditions</u> (flood+baseflow vs flushing). There is thus no repetition and we propose to keep both parts separately since reviewer 1 noted that the paper is "logically organized".	
lines 301-302: how do you explain this contrasting behavior?	This sentence in the "results" section was written to describe the observations detailed in the following lines. The explanation is given in the discussion section. However the sentence was deleted to clarify the paragraph.	
lines 370-374: how do you explain a different behavior of congeners of organic compounds (PAHs, PBCs)?	PAHs molecules and PCBs congeners are indeed two families of organic compounds, and it is well know that their sources are different (e.g. for PAHs: <a href="https://doi.org/10.1002/etc.5620160212">https://doi.org/10.1002/etc.5620160212</a> ). In the Rhône River, we highlighted such differences in a report of the OSR programm and concluded that the different congeners of PAHs have different origins, mainly road traffic and	For the different behavior of the PAHs congeners (Table S4), preliminary results on PAHs concentration in SPM collected in the upper Rhône River demonstrate that the different congeners of PAHs have different origins, mainly road traffic and domestic heating (Botha et al., 2014, Poulhier et al., 2018).

	<p>domestic heating. Moreover, the absence of relation between PAHs and PCBs demonstrate that PCBs mainly result from industrial releases. <a href="http://www.graie.org/osr/IMG/pdf/2018.01_livra_ble_iv2_vdiffusable_mc_gp_drm.pdf">http://www.graie.org/osr/IMG/pdf/2018.01_livra_ble_iv2_vdiffusable_mc_gp_drm.pdf</a></p> <p>Furthermore, the explanation about these differences is given in the “discussion” section at chapters 4.2 and 4.3 but more details were added to the manuscript.</p>	
<p>Discussion should not report citations of figures. Sub-titles in discussion are not needed</p>	<p>References to the figures in the discussion should help the reader to find which part of the results is related to this discussion. Also, sub-titles presented as questions were proposed in order to clarify the discussion.</p> <p>We are surprised by this reviewer comment, because almost all papers refer to figures in the discussion when necessary (see for examples: : <a href="https://doi.org/10.1016/j.jenvman.2019.109479">https://doi.org/10.1016/j.jenvman.2019.109479</a> or <a href="https://doi.org/10.1016/j.jenvman.2019.109391">https://doi.org/10.1016/j.jenvman.2019.109391</a> or <a href="https://doi.org/10.1016/j.jenvman.2019.109405">https://doi.org/10.1016/j.jenvman.2019.109405</a>).</p> <p>Based on the comment of reviewer 1 (“the paper is logically organized. I found hard to criticize the different sections”), we prefer to keep these citations and sub-titles.</p>	
<p>line 412: the day is missing in the date: "06/0/2012"?</p>	<p>Corrected</p>	<p>[...]from 06/11-14/2012 [...]</p>
<p>lines 419-422: any idea of contamination levels in sediments stored in the reservoirs?</p>	<p>Unfortunately investigations on contamination levels in the studied reservoirs are very sparse.</p>	

	<p>Concentration of TME, PAHs and PCBs measured on sediment cores collected in the studied dams were previously reported by the Forel Institute in 2009. However, each sediment cores were fully mixed and only one measure of contaminants were conducted on each mixed sediment cores. Therefore as our results demonstrate that the particle size affect the concentrations as well as the deepness of the sediment flushed, it is difficult to compare the results of the mixed sediment cores and our samples of SPM.</p>	
<p>lines 424 and 431: what do you mean with "origin"?</p>	<p>The meaning is explained in chapter 4.2 and 4.3. Origin means the source of particles that include soil of the catchments, sediments of the different tributaries, or anthropic sources (for example road traffic and domestic heating for PAHs).</p>	
<p>Paragraph 5: these are conclusions</p>	<p>We do not agree with this comment. Indeed, this section presents recommendations for scientists or stakeholders to improve monitoring of the consequences of flushing events in fluvial systems. These recommendations underline specifically how the monitoring of such events should differ from the one usually performed (e.g. regulatory monitoring) and could be implemented for all types of rivers.</p>	
<p>lines 482-458: this part of text is redundant. In particular: lines 482-521 could be moved to supplementary material, lines 522-548 could be deleted as they summarize results</p>	<p>We partly disagree with this comment. We rely on the reviewer1 comments to support this position. However, we shortened the text to avoid redundancy as much as possible.</p>	<p>The full manuscript was shortened as much as possible and some sections have also been moved to a new supplementary material section.</p>
<p>lines 496-500: also a pressure analysis in the watershed could drive the choice of contaminants of interest</p>	<p>Right. We add this point in the text.</p>	<p>Selection criteria include: priority pollutants according to regulations, performance of the analytical methods (limit of quantification, uncertainty) and emergent pollution, watershed</p>

		pressure;
Fig. 1: the city of Geneva is missing in the map	Corrected	
Figure 5: maybe both events should be plotted in the same graph (even if some points will be hidden behind the others): in the present form black points represent both normal conditions and one flushing event, and this creates some confusion	Corrected	
Tab. 1: why is the number of samples (n) different in each column of the same row?	This table was moved to the supplementary materials (Table S4) to shorten the manuscript. The number of samples was corrected and is now the same for each row of particle size.	

# Dam flushing operations



### Particulate contaminant concentrations

- PAHs
- 
- Phthalate
- PCBs
- ETMs
- Alkyl-phenols

## Highlights

- Dam flushing operations were monitored on the Upper Rhône River (France)
- Sediment dynamic differed during flushing operations and flood events
- About 0.6 Mt of SPM were stored in the Upper Rhône from 2011 to 2016
- 21 to 37% of the mean annual SPM flux transited during the 2012 flushing operations
- Particulate contaminant concentrations were driven by particle size and SPM origin

# 1 **Impact of dam flushing operations on sediment dynamics and** 2 **quality in the upper Rhône River, France**

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4 Gairoard<sup>c</sup>, Olivier Radakovitch<sup>a,c</sup>, Marina Coquery<sup>b</sup>

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## 14 **Abstract**

15 The Rhône River (France) has been used for energy production for decades and 21  
16 dams have been built. To avoid problems due to sediment storage, dam flushing operations  
17 are periodically organized. The impacts of such operations on suspended particulate matter  
18 (SPM) dynamics (resuspension and fluxes) and quality (physico-chemical characteristics and  
19 contamination), were investigated during a flushing operation performed in June 2012 on 3  
20 major dams from the Upper Rhône River. The concentrations of major hydrophobic organic  
21 contaminants (polychlorinated biphenyls, polycyclic aromatic hydrocarbons - PAHs, bis(2-



22 ethylhexyl)phthalate [DEHP] and 4-n-nonylphenol), trace metal elements, particulate organic  
23 carbon (POC) and particle size distribution were measured on SPM samples collected during  
24 this event as well as on those obtained from 2011 to 2016 on a permanent monitoring  
25 station (150 km downstream). This allows to compare the SPM and contaminant  
26 concentrations and fluxes during the 2012 dam flushing operations with those during flood  
27 events and baseflow regime. At equal water discharge, mean SPM concentrations during  
28 flushing were on average 6-8 times higher than during flood events recorded from 2011 to  
29 2016. While of short duration (19 days), the flushing operations led to the resuspension of  
30 SPM and contributed to a third of the mean annual SPM flux. The SPM contamination was  
31 generally lower during flushing than during baseflow or flood, probably due to the fact that  
32 flushing transport SPM only issued from resuspended sediment, with no autochthonous  
33 particles nor eroded soil. The only exception are PAHs and DEHP with higher concentrations  
34 during flushing, which must be issued from the resuspension of legacy-contaminated  
35 sediments stored behind the dams before the implementation of emission regulation.  
36 During flushing, the variations of POC and contaminant concentrations are also mostly  
37 driven by particle size. Finally, we propose a list of recommendations for the design of an  
38 adequate monitoring network to evaluate the impact of dam flushing operations on large  
39 river systems.

40

#### 41 **Key words**

42 Trace metals, PAHs, suspended particulate matters, pollution monitoring, sediment flux

## 43 **1. Introduction**

44 Investigating suspended particulate matter (SPM) transport in river catchments is a  
45 key component of the environmental monitoring of large rivers as SPM may cause important  
46 environmental impacts (Frings and Ten Brinke, 2018; Le Bissonnais *et al.*, 2005; Walling,  
47 2005). Suspended particulate matter transport can result in increasing water turbidity,  
48 degradation of fish habitat (Grimardias *et al.*, 2017), siltation of reservoirs (Owens *et al.*,  
49 2005), and transport of contaminants such as polychlorinated biphenyls (PCBs), trace metal  
50 elements (TME), radionuclides or nutrients (Dumas *et al.*, 2015; Evrard *et al.*, 2015;  
51 Horowitz, 2008; Taylor and Owens, 2009). The monitoring of SPM and associated particulate  
52 contaminant fluxes in rivers is therefore necessary, though practically challenging.

53 One of the challenges is to evaluate the role of dam reservoirs in terms of both sink  
54 and source of SPM (Hauer *et al.*, 2018). Indeed, dams are among the main structures that  
55 impact SPM transport by reducing flow velocity and increasing sediment deposition (Syvitski  
56 *et al.*, 2005). Such storage leads to a decrease in reservoir capacity and triggers economic  
57 issues (White, 2001), hence several methods are used to limit reservoir filling (Kondolf *et al.*,  
58 2014). Dam flushing operations are regularly organized to remove sediments stored in  
59 reservoirs and avoid problematic consequences such as siltation and enhanced flood hazard  
60 (Brown, 1944; Peteuil *et al.*, 2013). These operations consist in increasing water velocity and  
61 shear stress by lowering the water level to erode material deposited in the reservoir (Brandt,  
62 2000; Di Silvio, 2001; Kondolf *et al.*, 2014). However, flushing operations can have significant  
63 ecological impacts: increased stress or mechanical damage of organisms, loss of habitats,  
64 and decrease in dissolved oxygen concentrations due to increased SPM concentrations (Espa  
65 *et al.*, 2016; Grimardias *et al.*, 2017; Hauer *et al.*, 2018; Kemp *et al.*, 2011). Besides these

66 direct consequences, contaminants associated with deposited sediments (and legacy  
67 contamination) may also be resuspended and disseminated (Peter *et al.*, 2014) or  
68 transferred to the dissolved phase (Bretier *et al.*, 2019; Kalnejais *et al.*, 2010), increasing  
69 their bioavailability (e.g. Dong *et al.*, 2018 for TME). On the contrary, resuspension of less  
70 contaminated sediments may result in the dilution of particulate contamination (Ferrand *et*  
71 *al.*, 2012). While difficult to evaluate, especially in large river systems, these impacts should  
72 however be better addressed in order to minimize the costs under consideration of  
73 ecological requirements and to improve sediment management in terms of hydropower use  
74 (Hauer *et al.*, 2018).

75         The Rhône River is France's number one river in terms of water discharge and  
76 hydroelectricity production. Downstream of Lake Geneva in Switzerland, the Rhône River is  
77 highly artificialized and characterized by the presence of 21 hydropower plants. A large  
78 proportion of SPM is stored each year in this river, mostly in dam reservoirs (Bravard and  
79 Clémens, 2008; Provansal *et al.*, 2014). As several industries are implemented between Lake  
80 Geneva and the Rhône River Delta, a large range of contaminants is delivered to the river  
81 (e.g., Ollivier *et al.*, 2011) and a fraction is also trapped in dam reservoirs. Dam flushing  
82 operations are regularly conducted on the upper Rhône River to prevent bed aggradation in  
83 the Verbois reservoir (Figure 1), and thus urban flooding in the city of Geneva (Peteuil *et al.*,  
84 2013). From 1945 to 2003, flushing operations were performed every 3 years and the  
85 quantity of SPM exported was similar to the quantity of sediment stored since the previous  
86 flush (Diouf, 2013). Due to the associated increase of turbidity and potential consequences  
87 on fish populations (Grimardias *et al.*, 2017), such operations are now strictly monitored in  
88 order to reduce their environmental impact. However, the survey of SPM transport on a

89 large scale remains difficult and the impact of such operations compared to flood events is  
90 still a matter of debate due to the lack of data. Additionally, we still need a better  
91 understanding of contaminants remobilization and transport processes under such  
92 conditions (Hauer *et al.*, 2018; SedNet, 2014), as floods and dam flushing operations are  
93 major events able to transport a large fraction of contaminant fluxes (Poulier *et al.*, 2019).  
94 The behavior and dynamics of sediment (and associated contaminants) and dissolved/labile  
95 metals have already been studied on the Rhône River during floods and flushing operations,  
96 respectively (Antonelli *et al.*, 2008; Ollivier *et al.*, 2011; Sicre *et al.*, 2008; Bretier *et al.*, 2019;  
97 Peter *et al.*, 2014). However, there is a lack of information regarding the impact of flushing  
98 operations on particulate contaminants.

99         This study aimed at characterizing the impact of flushing operations on SPM  
100 dynamics and quality in a large river system: the Rhône River basin. Thanks to a specific  
101 monitoring, the fluxes and mass balances of SPM and associated contaminants triggered by  
102 flushing operations in 2012 and 2016 were estimated at different time scales. The variations  
103 in contaminant concentrations were related to the characteristics of SPM (particle size  
104 distribution, organic carbon content) as well as to their origin in the watershed (eroded soil  
105 versus resuspended sediment). Finally, the SPM monitoring performed during a second  
106 flushing operation in 2016 is also used to compare and evaluate the role of such events on a  
107 pluriannual scale.

## 108 2. Materials and Method

### 109 2.1. Location of the dams and description of the dam flushing operations

110 The Rhône River (95 600 km<sup>2</sup> watershed area, mean water discharge of ~1700 m<sup>3</sup>.s<sup>-1</sup>  
111 at the outlet station of Beaucaire) constitutes the main SPM input to the Western  
112 Mediterranean Sea, with a mean SPM flux of 6.5 Mt.year<sup>-1</sup> from 2000 to 2015 according to  
113 Poulhier *et al.* (2019). The Rhône Sediment Observatory operates two permanent monitoring  
114 stations along this river and several others on its main tributaries (Poulhier *et al.*, 2019;  
115 Thollet *et al.*, 2018). We used data collected at the permanent station of Jons (45.8121N,  
116 5.0896E – see Fig. 1), near the city of Lyon, to investigate the dynamics of sediments in the  
117 upper Rhône River and notably the impact of dam flushing operations conducted  
118 downstream of Lake Geneva.

119 This study focuses on flushing operations conducted in June 2012 in three reservoirs  
120 located downstream of Lake Geneva (Figure 1): Verbois, Chancy-Pougny, and Génissiat.  
121 Flushing operations used to be performed in this area every 3 years from 1945 to 2003, but  
122 the Swiss authorities later decided to consider alternative methods to manage SPM and  
123 dams. More than  $3 \times 10^6$  m<sup>3</sup> of SPM were thus stored in the Verbois reservoir between 2003  
124 and 2012 (Services industriels de Genève, 2014). They mainly originated from the Arve River,  
125 a tributary located upstream of the dam (Figure 1), which transports  $\sim 0.7 \times 10^6$  m<sup>3</sup> of SPM  
126 per year (approx. 1 to 3 tons dry weight), half of this amount being stored behind the dam  
127 (Guertault *et al.*, 2014; Launay *et al.*, 2019). This accumulation induced a change in the water  
128 level of the reservoir during floods that could impact the security of local population. Since  
129 no alternative management solutions were validated, the Swiss and French authorities

130 agreed to conduct a new flushing operation in June 2012 on the three successive dams  
131 (Figure 1). The flushing operations were conducted by lowering the water level following the  
132 drawdown flushing method (Fruchard and Camenen, 2012) in order to remove stored SPM  
133 in the three dams (Figure 2). The large-volume Génissiat reservoir located downstream of  
134 the two other dams (Figure 1) was flushed first (4-12 June) in order to compensate for the  
135 deposition of the SPM released from the Chancy-Pougny (9-16 June) and Verbois dams (9-22  
136 June, Figure 2; Diouf, 2013). Five small dams located between Génissiat and Jons (Figure 1)  
137 were also opened during this period and managed in order to prevent both sediment  
138 deposition and resuspension. The by-passed sections of the Rhône River (Old Rhône reaches)  
139 were disconnected and the whole SPM flux transiting through the reservoirs and tailrace  
140 canals. The levels of the reservoirs were lowered as much as possible to prevent deposition  
141 and ensure a quick transfer of water and SPM. It should also be noted that floods occurred in  
142 two upstream tributaries, Ain and Fier, during the 2012 flushing operations.

143 Another flushing operation, also monitored at Jons, was conducted on the same  
144 dams in May 2016 (Diouf, 2017). Compared to 2012, the 2016 flushing operation was only  
145 partial, with a smaller decrease of the water level in Verbois reservoir: 361 m a.s.l. in 2016 vs  
146 352 m a.s.l. in 2012 (Diouf, 2017). Contrary to 2012, the 3 dams were flushed at the same  
147 time and during 11 days (from May 20<sup>th</sup>, 2016 at 12:00 to May 31<sup>st</sup>, 2016 at 12:00).

## 148 **2.2. Water discharge**

149 There is no hydrometric station close to Jons and hourly water discharge was  
150 calculated using the 1-D hydrodynamical model MAGE (Irstea, France) and discharge inputs  
151 from upstream hydrometric stations (Lagnieu), Bourbre (Tignieu-Jameyzieu), and Ain (Port-  
152 Galland) Rivers (Figure 1) (Dugué *et al.*, 2015; Launay *et al.*, 2015). This model is calibrated

153 for the whole Rhône River and outputs were evaluated against measured water levels, with  
154 a maximal accepted difference of 10 cm.

### 155 **2.3. Concentration of SPM**

156 To evaluate the SPM concentration at Jons, turbidity measurements were conducted  
157 continuously every 15 min using a Hach Lange SC200 turbidity probe. The relation between  
158 turbidity (in NTU) and SPM concentration was calibrated as follows: SPM concentration was  
159 measured in water samples collected during various hydrological conditions (baseflow,  
160 flood, and flushing operations) in order to cover the entire range of discharges. Those water  
161 samples were collected manually or using a portable automatic water sampler (ISCO or  
162 SIGMA – 1 sample every 4 hours) due to logistic constrains. Suspended particulate matter  
163 concentration was measured after filtration of the samples through pre-weighed filters  
164 (Whatman GF/F, 0.7  $\mu\text{m}$ ). The limit of quantification (LQ) was 2  $\text{mg}\cdot\text{L}^{-1}$ .

### 165 **2.4. SPM sampling for chemical analyses**

166 In order to collect a sufficient amount of SPM material for subsequent  
167 physicochemical analyses, two sampling methods were used at Jons, Continuous Flow  
168 Centrifugation (CFC) and Particle Trap (PT) (Masson *et al.* 2018), as described in details in the  
169 supplementary materials (Supplementary Material #1.1). The CFC samples were used to  
170 characterize the concentration of TME (Cd, Cu, Pb, Zn), Hg and PCBs. The PT was used to  
171 collect a sufficient amount of SPM for chemical analysis of polycyclic aromatic hydrocarbons  
172 (PAHs), bis(2-ethylhexyl)phthalate (DEHP) and 4-n-nonylphenol. During the 2012 flushing  
173 operations, samples were collected every day from June 4 to June 16 with the CFC and

174 during 2-3 day periods from June 5 to June 16 with the PT (supplementary material #2 Table  
175 S1).

176 At the Jons station, the end of the pumping pipe is installed approximately 5 m  
177 downstream of the PT in order to side step potential heterogeneity of SPM throughout the  
178 river cross-section. Prior to chemical analysis, SPM collected with the two sampling  
179 techniques were transferred to clean brown glass bottles (250 mL), deep-frozen (-18°C),  
180 freeze-dried, and finally homogenized by grinding in an agate mortar. SPM samples were  
181 stored in the dark and at ambient temperature before analysis.

## 182 **2.5. Grain-size distribution and particulate organic carbon analysis**

183 Volumetric grain size distribution was analyzed on a CILAS 1190 laser particle size  
184 analyzer (range of values: 1 – 2 500  $\mu\text{m}$ ) according to the ISO 13320 standard method  
185 (AFNOR, 2009). Recent investigation on Particle Size Distributions (PSD) in the Rhône River  
186 concluded that the usual percentiles d10, d50 or d90 might not be sufficient to describe  
187 grain size due to a multimodal shape (Masson *et al.*, 2018). This shape can be modeled as a  
188 combination of several homogeneous subpopulations of mixed particles, each following a  
189 log-normal distribution. Therefore, each subpopulation was modeled using the R software  
190 (version R3.2.0) to extract the mass proportion of each subpopulation, which were used to  
191 compare the different PSD. Subpopulations were clustered in five classes according to their  
192 modal diameter: < 4  $\mu\text{m}$  (clay), 4-15  $\mu\text{m}$  (fine silt), 15 – 63  $\mu\text{m}$  (coarse silt), 63 - 125  $\mu\text{m}$  (very  
193 fine sand) and > 125  $\mu\text{m}$  (sand).

194 The determination of particulate organic carbon (POC) in SPM samples was  
195 performed using a carbon analyzer (Thermo Electron, CHN Flash 2000) at the INRA  
196 laboratory (Arras, France). Decarbonation was performed using hydrochloric acid



197 according to the NF ISO 10694 standard method (AFNOR, 1995). The LQ was estimated to be  
198  $50 \text{ mg.kg}^{-1}$  by using a reference material (Aglae, 15 M9.1;  $40 \text{ g.kg}^{-1}$ ) and the analytical  
199 uncertainty varied between  $\sim 3\%$  and  $\sim 6\%$  ( $k=2$ ), depending on the POC concentration.

## 200 **2.6.Chemical analysis of SPM**

201 Concentrations of total Cd, Cu, Pb, and Zn were determined after a microwave acid  
202 digestion of the SPM as described in the supplementary materials (Supplementary Material  
203 #1.2). The LQ was  $0.05 \text{ }\mu\text{g.kg}^{-1}$ . The mean difference between measured and certified values  
204 were, for STSD-3 ( $n=6$ ) and MESS-4 ( $n=3$ ), respectively: 20 and 3% for Pb; 7 and 6% for Cu; 5  
205 and 2% for Zn; 14 and 7% for Cd.

206 The determination of total Hg in SPM was performed using an automated atomic  
207 absorption spectrophotometer, DMA 80 (Milestone), according to EPA method 7473 (US  
208 EPA, 2007). The LQ was  $10 \text{ }\mu\text{g.kg}^{-1}$ . Blanks and certified reference materials (IAEA 433,  
209 marine sediment; LGC 6187, river sediment) were systematically used to check analytical  
210 accuracy (94%) and uncertainty (14%;  $k=2$ ).

211 Indicator PCBs (PCB 28, 52, 101, 118, 138, 153, and 180) were analyzed using capillary  
212 gas chromatography coupled to an electron capture detector (GC-ECD), as detailed in  
213 Masson *et al.* (2018). Limits of quantification ranged from 0.5 to  $1 \text{ }\mu\text{g.kg}^{-1}$ . Only results for  
214 compounds with a quantification frequency higher than 60% (Helsel, 2006) were considered  
215 here, i.e. PCBs 101, 138, 153, and 180.

216 The 16 priority PAHs (Keith, 2015) were analyzed by the Laboratory of Hygiene and  
217 Environment (Rouen, France), using capillary gas chromatography coupled to a mass  
218 spectrometer (GC-MS) and according to the XP X33-012 Standard (AFNOR, 2000). Limits of  
219 quantification ranged from 1 to  $2 \text{ }\mu\text{g.kg}^{-1}$  for most of the PAHs, except for

220 dibenz[a,h]anthracene and fluorene (up to 5  $\mu\text{g.kg}^{-1}$ ) and acenaphthylene (up to 20  $\mu\text{g.kg}^{-1}$ ). In  
221 this paper, the 16 PAHs are presented and discussed as their quantification frequency was  
222 higher than 60%.

223 Alkylphenols (4-n-nonylphenol, octylphenol, para-tert-octylphenol, tert-butylphenol)  
224 and DEHP were analyzed by the La Drôme laboratory (Valence, France), by GC-MS. Limits of  
225 quantification were 10  $\mu\text{g.kg}^{-1}$  for alkylphenols and 100  $\mu\text{g.kg}^{-1}$  for DEHP. In this paper, only  
226 substances with a quantification frequency higher than 60% are presented, i.e 4-n-  
227 nonylphenol and DEHP. Analytical uncertainties of these organic contaminants were  
228 estimated at 60% ( $k=2$ ) for concentrations lower than 3-times the LQ and 30% ( $k=2$ ) for  
229 concentrations higher than 3-times the LQ. For concentrations higher than 3-times the LQ,  
230 this value was confirmed by interlaboratory trials (Charpentier, 2016).

## 231 **2.7. SPM and contaminant fluxes calculation**

232 One important question regarding the impact of flushing operations is to determine  
233 their relative contribution to SPM transport compared to flood events. Such a comparison  
234 requires an estimation of the influence of both types of events over a longer time scale. The  
235 Jons station allows an estimation of the annual SPM fluxes from 2011 to 2016 (based on  
236 hydrological years, i.e. from September to August). The SPM fluxes were calculated from the  
237 mean hourly discharge and SPM concentration time series and cumulated over periods of  
238 flood events, flushing operations, or years (Poulier *et al.*, 2019). Contaminant fluxes were  
239 calculated by multiplying the hourly SPM flux by the particulate contaminant concentrations.  
240 Station-specific median particulate contaminant concentrations were used for the non-  
241 monitored periods to fill gaps in the measured time series (Poulier *et al.*, 2019). The BDOH

242 tool was used to do the calculation of SPM and contaminant fluxes (Branger *et al.*, 2014),  
243 and data are available online (Thollet *et al.*, 2018).

## 244 **2.8. Classification according to hydrological conditions**

245 In order to investigate the impact of flushing operations, results were compared to  
246 baseflow and flood conditions using data collected at Jons from 2011 to 2016. Samples  
247 classified as “flushing period” refer to those collected between 06/04/2012 00:00 and  
248 06/22/2012 15:00. Samples were classified as “flood samples” if the corresponding discharge  
249 was higher than a  $800 \text{ m}^3 \cdot \text{s}^{-1}$  discharge threshold (i.e., half the 2-year flood discharge  
250 (Launay, 2014)). For time-integrative samples collected with a PT, classification was based on  
251 the SPM proportion related to the different hydrological states. For each PT sample, the  
252 corresponding SPM flux was calculated using turbidity values for the entire period of  
253 collection (total SPM flux) and for the period with water discharge higher than the flood  
254 threshold (SPM flux during flood). The PT samples were classified as flood samples if the  
255 SPM flux during the flood event represented more than 50% of the total SPM flux. The other  
256 samples were classified as “baseflow samples”.

## 257 **3. Results**

### 258 **3.1. Variation of water discharge, SPM concentration and** 259 **quality during flushing operations**

260 At Jons, the discharge ranged from 343 to  $1600 \text{ m}^3 \cdot \text{s}^{-1}$  (maximum reached on June 14)  
261 during the 2012 flushing operations, while SPM concentration ranged from 10 to  $905 \text{ mg} \cdot \text{L}^{-1}$   
262 (maximum reached on June 13) (Figure 3). The POC concentration was  $13.5 \pm 3.8 \text{ mg} \cdot \text{kg}^{-1}$

263 (mean  $\pm$  standard deviation) but varied depending on the sampling method (see  
264 supplementary material #2 Table S1).

265 The PSD were mostly characterized by silt (fine silt (from 30 to 82%), coarse silt (5%-  
266 70%)) and clay (0-34%, supplementary material #2 Table S1) depending on the sampling  
267 method (see supplementary materials #1.3). Sand particles were only observed in two  
268 samples and the proportion never exceeded 26% (supplementary material #2 Table S1).

269 The TME concentrations remained fairly stable during all the flushing period, while a  
270 decrease of the levels of Hg and PCBs was observed in the last collected sample (Figure 4 –  
271 supplementary material #2 Table S2). For all the PAHs and 4-n-nonylphenol, the  
272 concentration of the first samples (collected from June 7 to 11) were at least 1.2 times  
273 higher (up to 6 times) than the two other samples (collected from June 11 to 16) (Figure 4 –  
274 supplementary material #2 Table S3). For DEHP, the only clear trend is a lower value (by a  
275 factor ten) found in the first sample.

## 276 **3.2. Specificity of SPM characteristics during flushing events**

### 277 **3.2.1. Water discharge and SPM concentration**

278 The water discharge measured at Jons during the 2012 flushing operations was  
279 similar to that of several floods occurring that year (Figure 3). Peaks of SPM concentration  
280 were concomitant to those of water discharge (June 13<sup>th</sup> and 14<sup>th</sup>), and the highest one  
281 during the flushing operation was almost 25% higher than during flood events (Figure 3).  
282 Due to sediment resuspension in reservoirs, for equivalent water discharge at Jons, mean  
283 SPM concentrations during the flushing operations of June 2012 were 8 times higher than  
284 during floods (Figure 5). In 2016, the discharge was lower than in 2012 (Figure 5) due to a

285 different management of the flushing operations (Diouf, 2017). However, the mean SPM  
286 concentration was 6 times higher than during flood events at equivalent water discharge.

### 287 **3.2.2. SPM Fluxes**

288 From 2011 to 2016, the annual SPM fluxes (“output” fluxes) at Jons remained fairly  
289 constant with values ranging from 0.71 to 0.96 Mt.y<sup>-1</sup>. Flood events, representing 16-37% of  
290 the year duration, contributed to the major part of annual SPM fluxes and transported 48-  
291 89% of the annual flux (Figure 6). While representing only 3-5% of the year (Figure 6), the  
292 contributions of 2012 and 2016 flushing operations represented 37% and 23% of the total  
293 annual SPM flux, respectively. However, the contribution in 2012 might have been  
294 overestimated due to a flood event occurring in two upstream tributaries (Ain and Fier)  
295 during the flushing operation of Verbois dam. Thus, the contribution of the 2012 event  
296 should range from 0.15 to 0.26 Mt (21-37% of the annual flux) (Launay, 2014). Lastly, SPM  
297 fluxes during baseflow conditions remained quite constant over the years and contributed to  
298 less than 20% of the total annual fluxes.

299 The water discharges and SPM concentrations measured at other monitoring stations  
300 located upstream, on the main tributaries, were used to estimate the “input” SPM fluxes at  
301 Jons (Poulier *et al.*, 2019) (Figure 7). The two hydrological years with flushing operations  
302 (2012 and 2016) were characterized by higher output SPM fluxes compared to estimated  
303 input ones, with an export of about 0.1-0.2 Mt. In contrast, sediment storage of  
304 approximately 0.2 Mt.y<sup>-1</sup> was observed during years without flushing operations and despite  
305 the occurrence of flood events (outputs < inputs). From September 2012 to August 2016 (i.e.  
306 the period starting from the end of a flushing operation to the end of the next one), about  
307 0.6 Mt of SPM were thus stored in the Upper Rhône according to this estimation.

### 3.3. Relative contribution of flushing events to SPM and contaminant concentrations and fluxes

Contaminant concentrations, PSD and POC of samples collected during the 2012 flushing operations were compared to 2011-2016 data during other hydrological conditions (baseflow and flood) (Thollet *et al.*, 2018). Regardless of the sampling period or sampling method used, SPM were mostly silt sized with a non-negligible proportion of clay (Table S4). Presence of sand remained very sparse. The POC concentrations were almost twice lower in samples collected during flushing operations than for the other periods ( $p < 0.01$ , Table S4).

Different trends were observed for the contaminants of interest. Concentrations were not statistically different during flushing operations, baseflow, and flood conditions for Cd, 4-n-nonylphenol (Figure 8), PCB118 and PCB153 (supplementary material #2 Table S5). For the other TME (Cu, Pb, Zn) and for PCB101 and PCB138, concentrations during flushing operations were significantly lower ( $p < 0.05$ ) than under baseflow regime but similar to values measured during flood events (Figure 8, Table S5). For Hg, the mean concentration during flushing operations ( $39 \pm 6 \mu\text{g.kg}^{-1}$ ) was significantly lower than for other hydrological conditions ( $58 \pm 15 \mu\text{g.kg}^{-1}$  for baseflow and  $50 \pm 12 \mu\text{g.kg}^{-1}$  for floods). PCB180 exhibited a similar behavior (Table S5). In contrast, SPM sampled during flushing operations were more contaminated by PAHs and DEHP than SPM sampled during baseflow or flood conditions (Figure 8). Due to a low number of samples ( $n=3-4$ ), this increase was only significant ( $p < 0.05$ ) for some of the PAHs including fluorene (Figure 8, Table S5).

The fluxes of particulate contaminants during the June 2012 flushing operations were estimated based on discharge and SPM concentrations (Table 1). For all contaminants, the flushing operations transported about one third of the total annual flux.

## 331 4. Discussion

### 332 4.1. *Why are SPM concentrations higher during flushing* 333 *operations than during flood events at equivalent water* 334 *discharge?*

335 Our results demonstrate that the hydrological processes that occur during flushing  
336 operations are different from those occurring during flood events (Figures 5-7). In fact, the  
337 operations conducted on the dams by drawdown flushing to erode buried materials  
338 (Fruchard and Camenen, 2012) resuspended sediments that cannot be affected by flood  
339 events. This is why SPM concentrations were higher during flushing operations than flood  
340 events at equivalent water discharge. Thus, the use of a power relationship between  
341 discharge and SPM as described in Sadaoui *et al.* (2016) and Poulhier *et al.* (2019) would lead  
342 to the underestimation of the SPM concentrations and fluxes transiting during flushing  
343 operations (Figure 5). For example, the total SPM fluxes during the 2012 and 2016 flushing  
344 operations would be 0.07 Mt and 0.05 Mt, respectively, when using the equation proposed  
345 by Poulhier *et al.* (2019), i.e., almost 4 times lower than the measured fluxes (0.26 Mt and  
346 0.19 Mt, respectively – Figure 7).

347 The excess of annual SPM flux measured at Jons during hydrological years with  
348 flushing operations is related to the resuspension of sediment stored during years without  
349 flushing (Figure 7). The flushing operations led to a substantial resuspension of silt-sized  
350 particles that were stored during other flood events (Table S4). However, this estimation  
351 indicates a disequilibrium in the SPM budget over the six years studied on the Upper Rhône  
352 River (2011 – 2016 – Figure 7). Therefore, part of the stored sediments was not sufficiently

353 resuspended during flushing operations to reach Jons, and an excess of 0.6 Mt of sediment  
354 was trapped in this area despite the 2012 and 2016 flushing operations. Additional  
355 investigation on the reservoirs of this area would be necessary to characterise the  
356 proportion of SPM stored and resuspended during extreme events.

#### 357 **4.2. Which parameters influence particulate contaminant** 358 **concentrations?**

359 Variations of particulate contaminant concentrations were observed both during the  
360 flushing operations and during the different hydrological conditions.

361 First, the variations observed during the flushing operations for POC and several  
362 contaminant concentrations (Hg, PCBs; PAHs, 4-n-nonylphenol and DEHP) are mostly related  
363 to the PSD. The lowest concentrations of POC, PAHs and 4-n-nonylphenol were observed on  
364 samples collected from 06/11-14/2012 and characterized by at least two third of particles  
365 coarser than 15  $\mu\text{m}$  (coarse silt and sand). Also, the highest concentration of POC ( $22.1 \pm 0.9$   
366  $\text{mg}\cdot\text{kg}^{-1}$ ) was observed in the sample collected from 06/07-09/2012 with the second highest  
367 proportion of clay (17.5%). The increase in PAHs concentrations observed in the sample  
368 collected from the 06/09-11/2012 (Figure 4) was related to the resuspension of buried  
369 materials in the two most upstream dams as the operations started on 06/09/2012 at  
370 Verbois and Chancy-Pougny (Figure 2). This will be further discussed in the next section.

371 Second, the differences of POC and contaminants observed between the various  
372 hydrological conditions (Figure 8, Table S5) are related both to the particle size and the  
373 origin of the SPM. The decrease of concentrations observed during the flushing operations  
374 for most TME, Hg, PCB101, PCB138, and PCB150 is related to the PSD as such contaminants



375 are mainly adsorbed onto finest particles (Delle Site, 2001; Luoma and Rainbow, 2008; Steen  
376 *et al.*, 1978). In fact, coarser particles were transported during the flushing operations than  
377 during flood events or baseflow (Table S4). Decrease of POC due to PSD was observed both  
378 on CFC and PT samples. However, the presence of different trends depending on the  
379 contaminant demonstrates that other parameters such as the origin of the particles or local  
380 hot spots might also be involved. In this area, input by tributaries might be characterized by  
381 different geochemical signature as the concentrations observed for TME and Hg were  
382 associated with the geological backgrounds of their watersheds (Poulier *et al.*, 2019; Thollet  
383 *et al.*, 2018). For example, the mean concentration of Hg measured from 2011 to 2016  
384 varied by a factor 5 between the Arve River (main SPM contributor –  $0.026 \pm 0.008 \text{ mg.kg}^{-1}$ ,  $n$   
385 = 16) and the Bourbre River (highest Hg concentration -  $0.136 \pm 0.040 \text{ mg.kg}^{-1}$ ,  $n = 6$ ). For the  
386 different behavior of the PAHs congeners (Table S5), preliminary results on PAHs  
387 concentration in SPM collected in the upper Rhône River demonstrate that the different  
388 congeners of PAHs have different origins, mainly road traffic and domestic heating (Botha *et*  
389 *al.*, 2014, Poulier *et al.*, 2018).

390 Furthermore, preliminary work on modeling and fingerprinting conducted on SPM at  
391 Jons during the 2012 flushing operations confirmed that SPM mostly originated from the  
392 resuspension of SPM deposited in the dams (i.e. mainly from the Arve River) with a low  
393 contribution of particles freshly eroded from the other tributaries (Begorre *et al.*, 2018).

394 Finally, our results confirmed that despite the observed variation of concentrations  
395 on the Upper Rhône River, contaminant fluxes transported during dam flushing operations  
396 were mostly driven by an increase of SPM concentration, rather than by a change in  
397 contaminant contents.

398        **4.3.        *Can we explain the increase of some particulate contaminant***  
399                    ***concentrations?***

400            The observed increase of PAHs and DEHP concentrations during the flushing  
401 operations compared to other hydrological conditions (Figure 8 – supplementary material #2  
402 Table S5) may be related to the resuspension of legacy-contaminated sediments stored  
403 behind the dams, due to the drawdown flushing method used. In fact, the SPM resuspended  
404 and collected during the flushing operations were stored since 2003 (the last previous  
405 flushing operation), i.e. before the implementation of emission regulation of these  
406 pollutants (EC, 2008). On the other hand, SPM collected at Jons during baseflow and flood  
407 events started in 2011, i.e. after the implementation of the various regulations. PAHs are  
408 known to be produced mainly by incomplete combustion (pyrogenic processes) in domestic  
409 heating systems using wood (Hedberg *et al.*, 2002; Sicre *et al.*, 2008; Zhang and Tao, 2009).  
410 This induces higher PAH concentrations in urban areas and in winter. As reducing PAH  
411 emissions by 30% was a priority in the 2nd French National Health and Environment Plan  
412 (PNSE 2, 2009), the use of wood was gradually replaced by other more efficient heating  
413 systems. This was especially the case in the Arve basin (Figure 1), with a decrease of PAH  
414 emissions observed since 2008 (Atmo Auvergne-Rhône-Alpes, 2018). In fact, concentrations  
415 of Benzo[a]pyrene measured in SPM downstream of Chancy-Pougny from 2003 to 2008  
416 (NAIADES, 2019) ranged from 10 to 120  $\mu\text{g.kg}^{-1}$  ( $48 \pm 34 \mu\text{g.kg}^{-1}$ , n=24). In 2009,  
417 concentrations dropped and ranged from 16 to 67  $\mu\text{g.kg}^{-1}$  ( $38 \pm 25 \mu\text{g.kg}^{-1}$ , n=4). During the  
418 2012 flushing operations, the highest concentration ( $143 \mu\text{g.kg}^{-1}$ ) was measured on the SPM  
419 sample collected at the beginning of the operations on the most upstream dam (06/09/2012  
420 - Figure 1), and is therefore probably related to sediments stored before the regulation.

421 Finally, the increase of DEHP can also likely be explained by a change in regulations.  
422 In fact, DEHP is listed as a priority substance under the European Water Framework Directive  
423 (WFD; CEC, 2000). Its emissions are therefore regulated since 2008. DEHP was also included  
424 in 2011 in the Annex XIV of REACH by Regulation (EU, 2011), inducing the need of  
425 authorizations (and restrictions) for its specific uses.

## 426 **5. Feedback for future studies on sediment dynamics and quality** 427 **during dam flushing operations**

428 Our study demonstrates that SPM dynamics on the Upper Rhône River differed  
429 between flushing operations and natural floods, and confirms the necessity to monitor both  
430 discharge and SPM concentrations during such events. Also, the physico-chemical  
431 characteristics of SPM and associated contaminants might help to understand the dynamics  
432 and quality of the sediments. In this part, we focus on the best methodology to study  
433 sediment dynamics and quality during dam flushing operations.

### 434 **i) Design the monitoring to assess SPM concentrations and associated fluxes**

435 Both aspects (concentrations and fluxes) are important for assessing the ecological  
436 impact of dam reservoir flushing operations:

- 437 • For computing fluxes and mass budgets, continuous records of discharge and  
438 SPM concentration are required using gauging stations and turbidity records  
439 calibrated with frequent water samples. General relationships between SPM and  
440 discharge should not be used during flushing operations (see Figure 5).
- 441 • Monitor the entire flushing period for discharge and calibrated turbidity time  
442 series. We recommend assessing the type of flushing operations conducted (e.g.,

443 pressure flushing or drawdown flushing (Fruchard and Camenen, 2012; Kondolf *et*  
444 *al.*, 2014)).

445 **ii) Select the contaminants and other physico-chemical parameters to assess SPM**  
446 **quality**

- 447 • Optimise the selection of contaminants based on the results of previous  
448 monitoring (baseflow, flood, dam flushing operations or on sediments stored  
449 behind dam reservoirs), including the screening of new contaminants that were  
450 not sought or detected before;
- 451 • Selection criteria include: priority pollutants according to regulations,  
452 performance of the analytical methods (limit of quantification, uncertainty) and  
453 emergent pollution, watershed anthropic pressure;
- 454 • Monitor SPM quality over extended periods of time. Time integrative sampling  
455 systems like passive particle traps are appropriate (Masson *et al.*, 2018).

456 **iii) Extend the observations beyond the dam flushing event (temporally and**  
457 **spatially)**

458 Comparison with other periods of time and various hydrological conditions (average,  
459 baseflow, floods, other dam flushing events) are necessary to assess the relative impact of a  
460 dam flushing operation:

- 461 • It is useful to have at least one monitoring station with multi-year records of  
462 discharge, SPM concentration and physico-chemical parameters (POC and PSD);
- 463 • It is also important to monitor the discharge and SPM load of the main tributaries  
464 in the river system; additional information such as physico-chemical parameters

465 of the tributaries might also help understand the results observed at the  
466 monitoring station.

- 467 • Store SPM samples in proper conditions and with associated documentation to  
468 allow for future analyses, in case suspicious results are obtained for a given  
469 contaminant, or for future re-analysis of past events, for other contaminant  
470 and/or using better analytical methods.

#### 471 **4. Conclusions**

472 Variations of SPM and associated contaminant concentrations and fluxes were  
473 investigated from 2011 to 2016, including two dam flushing operations, at a permanent  
474 monitoring station located on the Upper Rhône River, France.

475 Despite the fact that flood events contributed to most of the annual flux of SPM,  
476 fluxes triggered by flushing operations were substantial (21-37% of the total annual SPM)  
477 despite their low annual duration (less than twenty days). These novel results demonstrate  
478 the necessity to have a long-term monitoring station located downstream to evaluate the  
479 impact of these operations.

480 Additional measurements of water discharge and SPM concentration along the Upper  
481 Rhône River and the main tributaries were used to investigate the spatial variation and to  
482 estimate the input/output SPM fluxes. The output fluxes measured at Jons were lower than  
483 the input fluxes during hydrological years without flushing operations and otherwise higher,  
484 attesting of a removal of the stored SPM. Flushing operations triggered the resuspension of  
485 sediments stored during flood events in reservoirs located along the Upper Rhône River. It is  
486 therefore crucial to continue such monitoring to investigate the fate of this excess of stored

487 sediment. Moreover, the composition of SPM changed during flushing operations as  
488 demonstrated by the analysis of several parameters and contaminants. Although the  
489 variations of POC and contaminants during the flushing operations were mostly related to  
490 changes in particle size, the origin of particles (e.g., resuspension of deeply stored  
491 sediments) was also important. Finally, we shared a general methodology to conduct similar  
492 monitoring.

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## Figure caption

Figure 1 – Location of the studied dams in the Upper Rhône River catchment with the main tributaries and the permanent monitoring station at Jons.

Figure 2 – Water level variation in the three dams studied on the Upper Rhône River during the 2012 dam flushing operations (modified from CNR, 2014).

Figure 3 – Variation of water discharge and the SPM concentration measured at Jons during the hydrological year of the studied period (September 2011 to August 2012). Flood threshold was estimated at  $800 \text{ m}^3 \cdot \text{s}^{-1}$ . The gray area represents the period of the 2012 flushing operations.

Figure 4 – Particulate contaminant concentrations ( $\pm$  analytical uncertainty) in SPM samples collected in the Rhône River at Jons during the flushing operations in June 2012.

Figure 5 – Relation between discharge and SPM concentration measured in the Rhône River at Jons from September 2011 to August 2016 (black circles). Data for the flushing operations of June 2012 (purple circle) and June 2016 (cyan circle) are shown. Flood threshold was estimated at  $800 \text{ m}^3 \cdot \text{s}^{-1}$ . The pink line represents the relationship between hourly water discharges and hourly SPM concentration at the station of Jons according to Poulhier et al. (2019).

Figure 6 - Proportion of annual SPM fluxes of the Rhône at Jons for the different hydrological conditions (baseflow, flood and flushing event) against the annual duration of the events

Figure 7 – Annual SPM fluxes of the Rhône River at Jons from 2011 to 2016 (hydrological years) for the different hydrological conditions (baseflow, flood and flushing events of 2012 and 2016); total output SPM fluxes (measured at Jons) and input SPM fluxes (computed from data on tributaries) are indicated for comparison.

Figure 8 – Particulate contaminant concentrations in SPM samples collected in the Rhône River at Jons manually, by CFC or by PT between hydrological conditions (baseflow, flood and flushing). For Cd, one outlier is not displayed for baseflow (value =  $2.41 \text{ mg} \cdot \text{kg}^{-1}$ ). Only the significant differences are displayed with \* for p value < 0.05, \*\* for p value < 0.01, \*\*\* for p value < 0.001 and \*\*\*\* for p value < 0.0001. Box plots represent the median and quartile values, Black circles and red diamonds represent outliers and mean values, respectively. n = number of samples.



Figure 1

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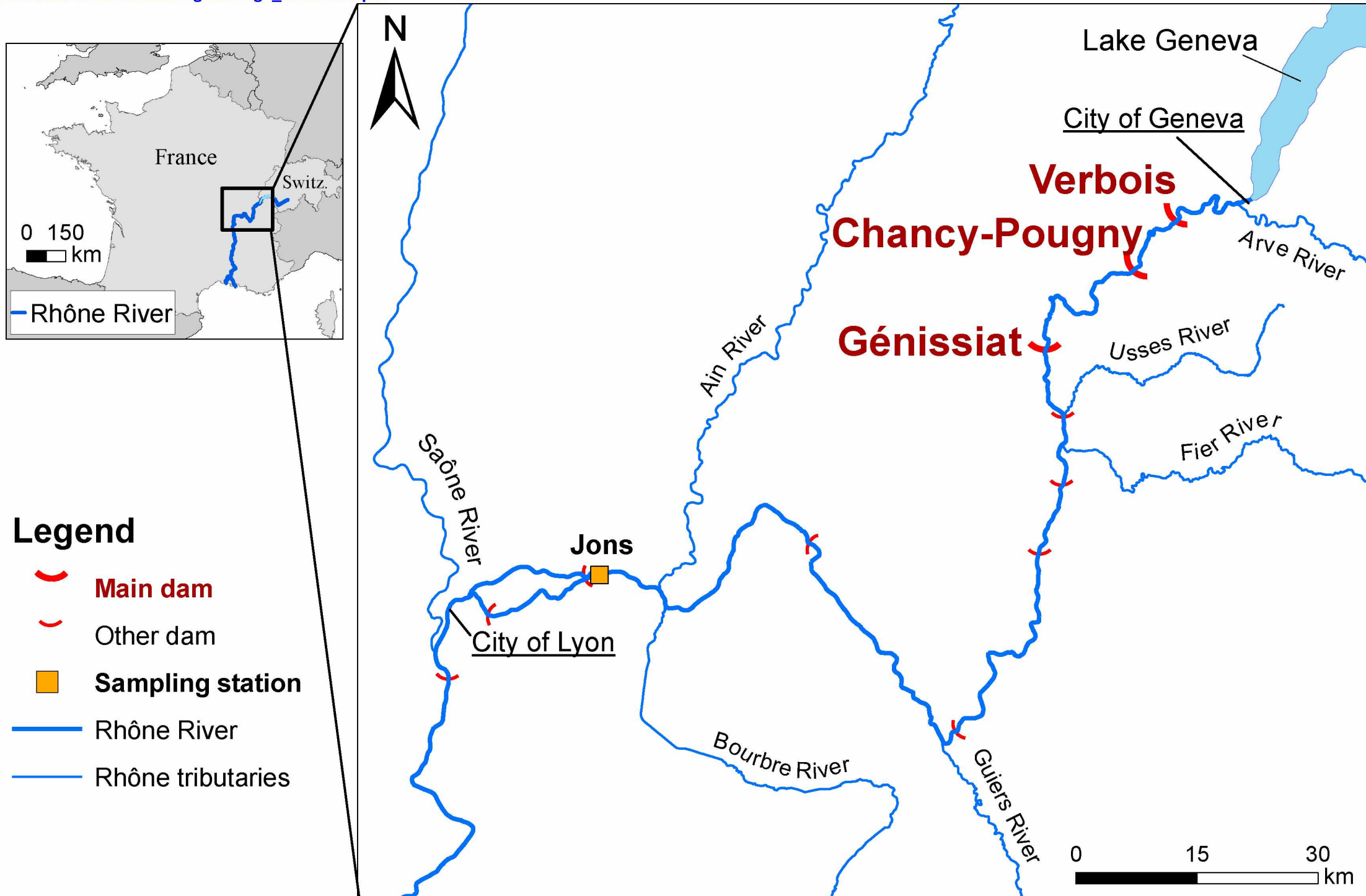
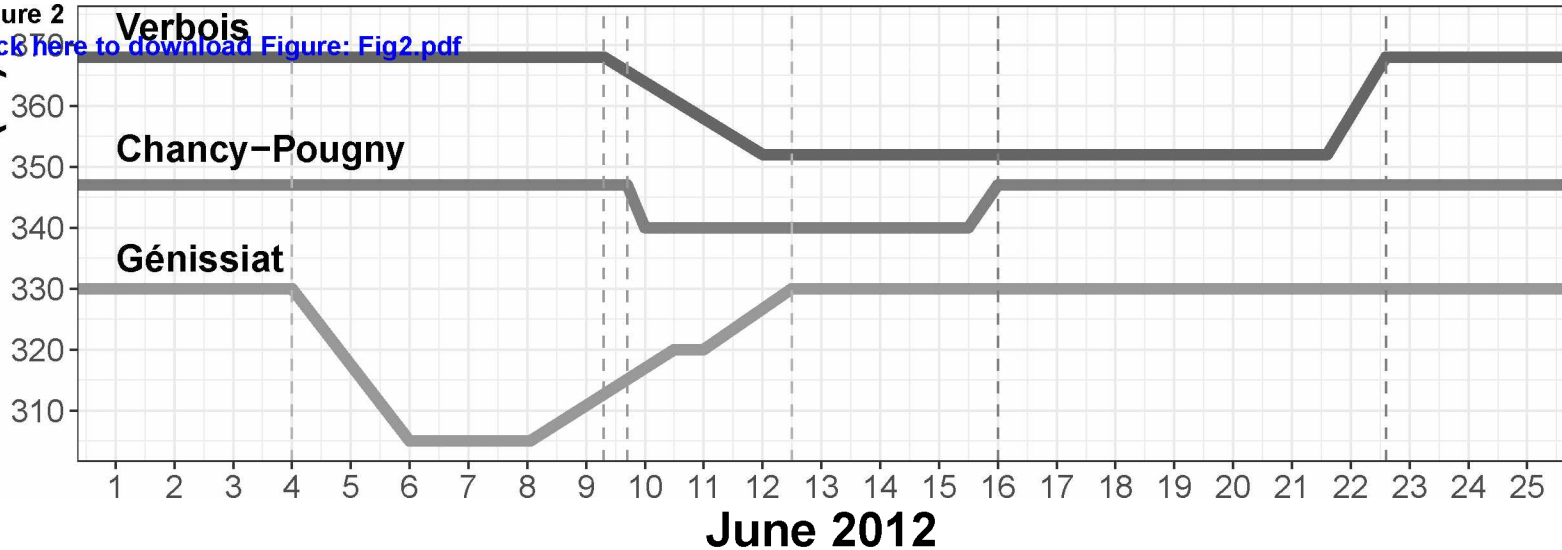
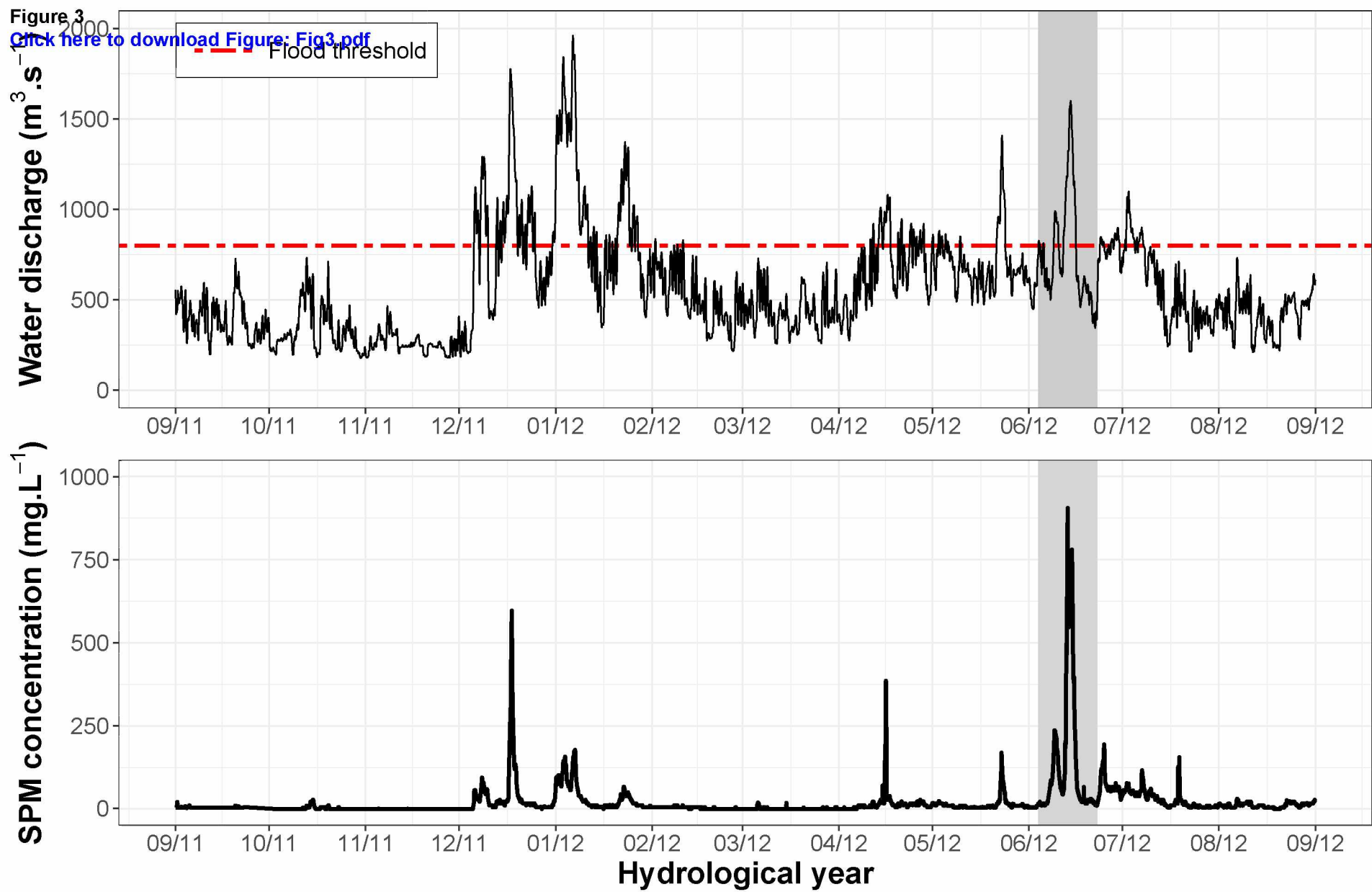


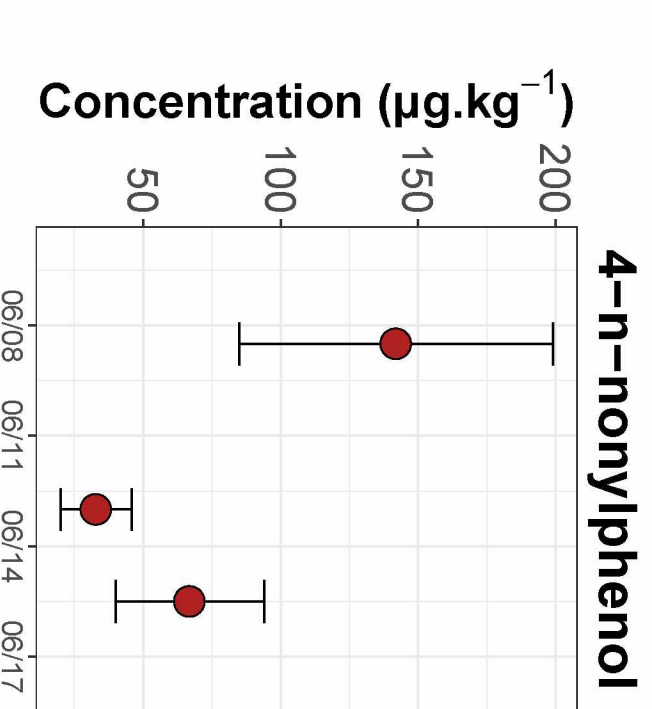
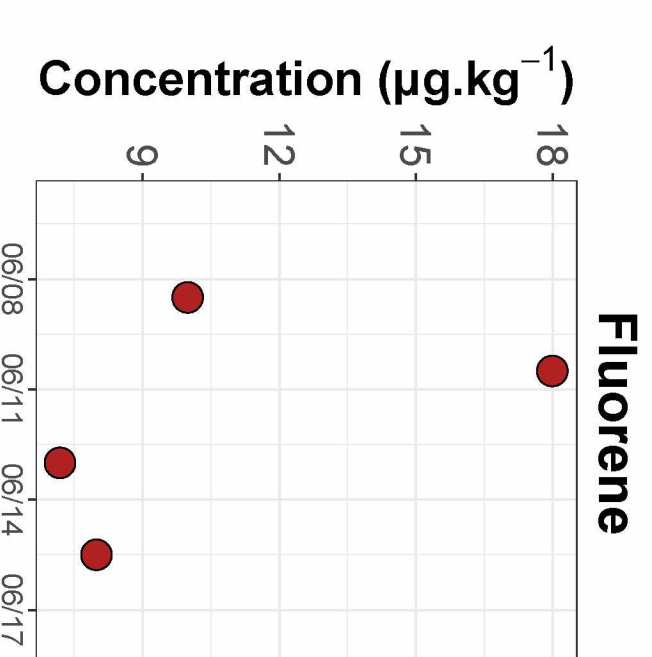
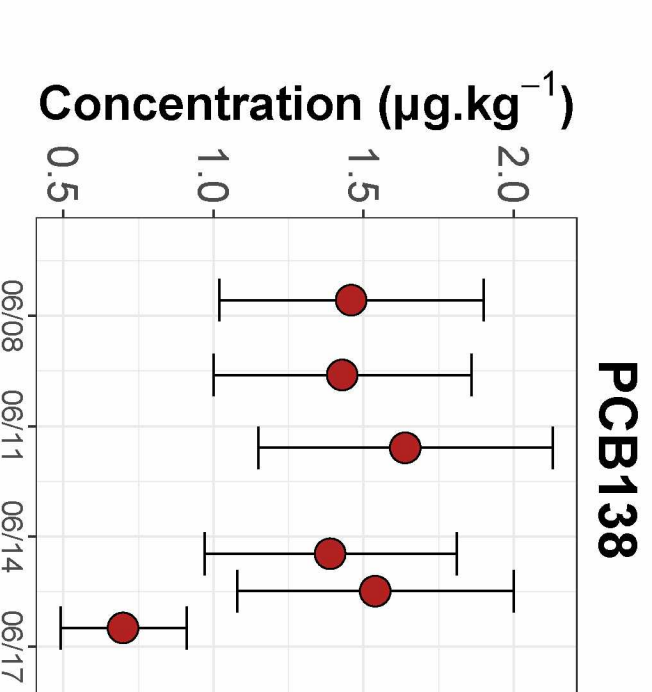
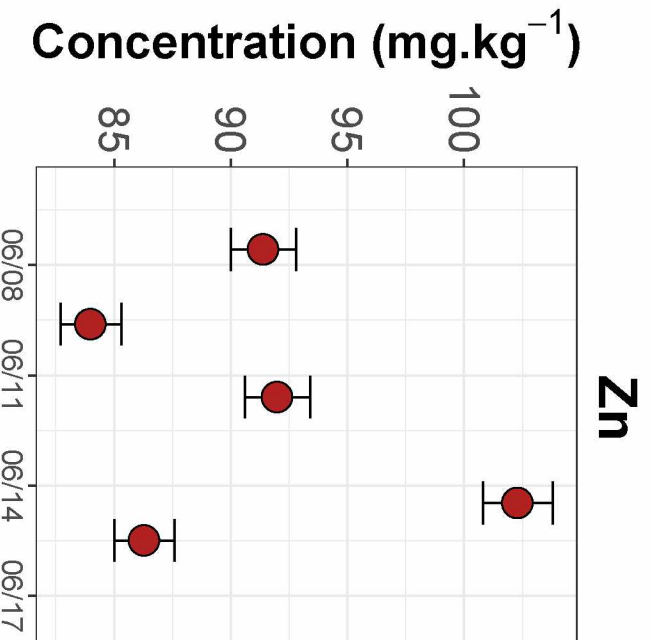
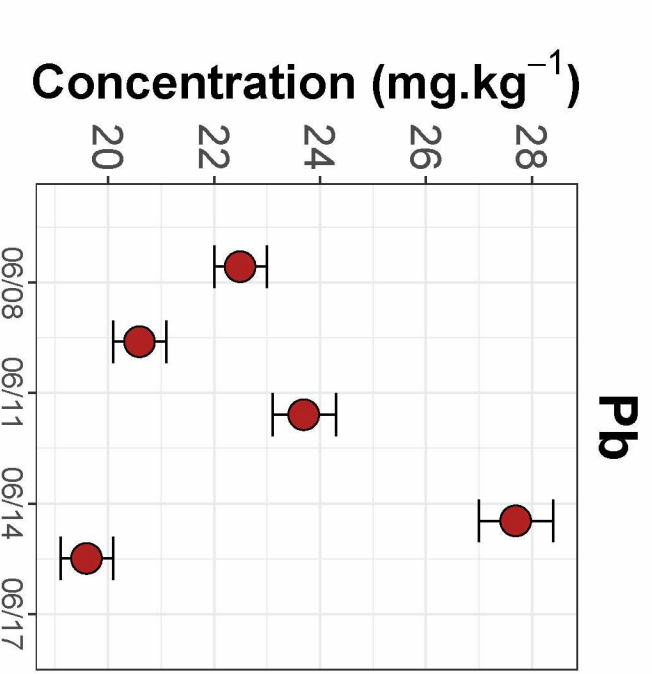
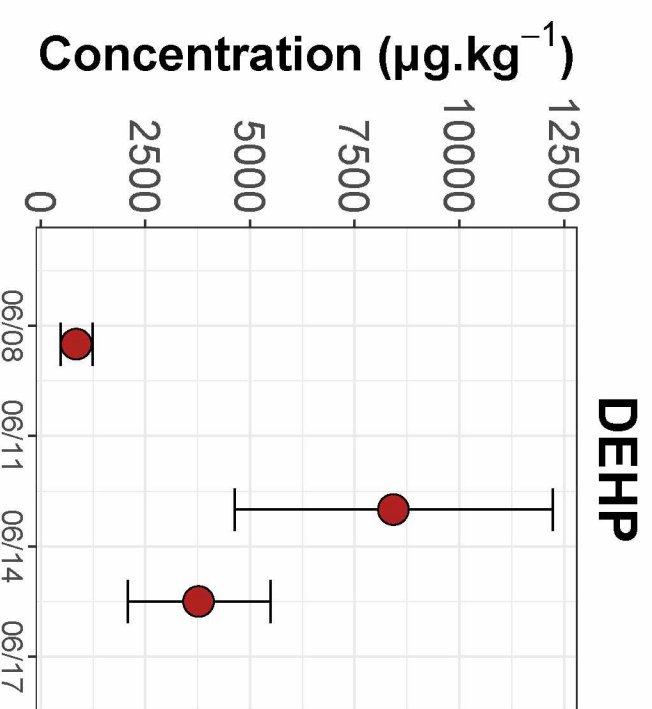
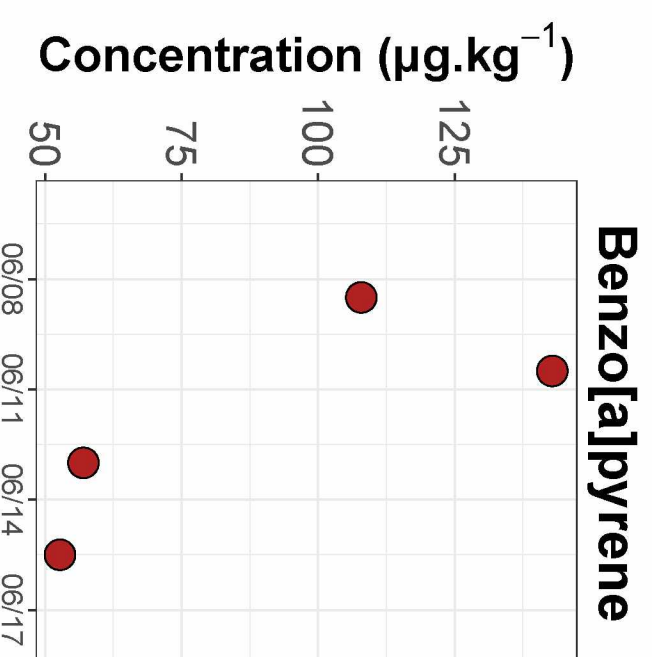
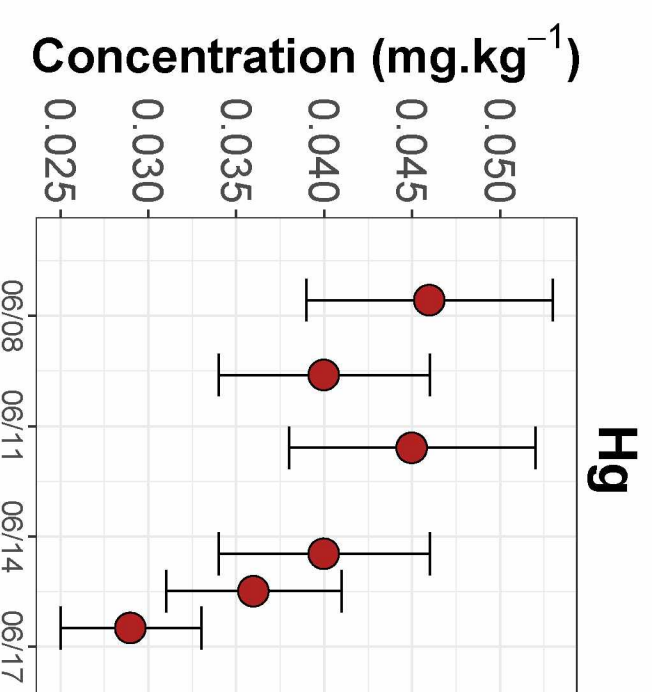
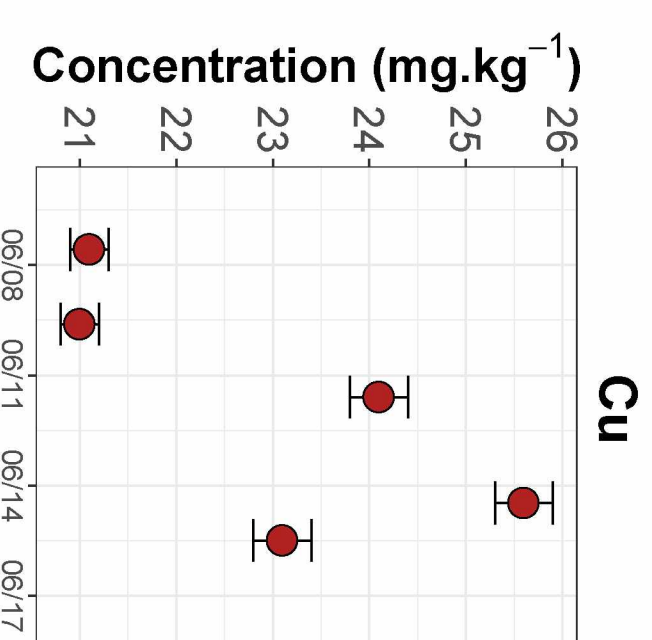
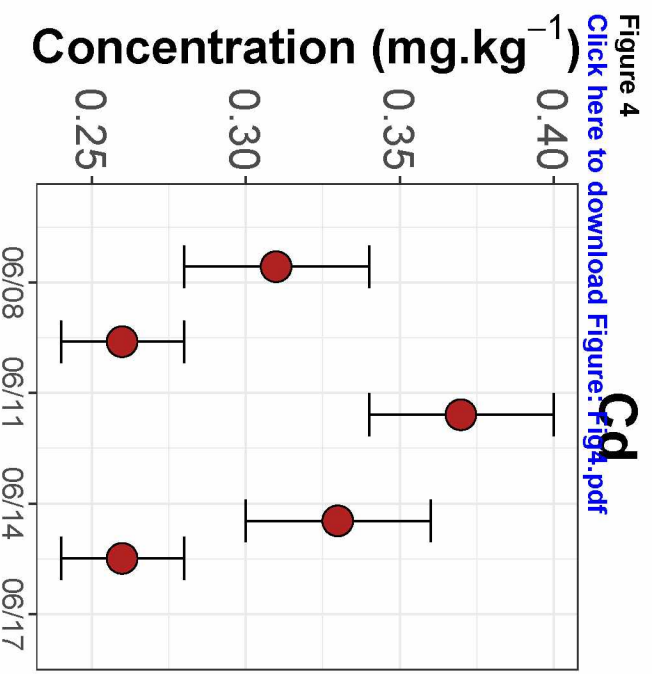
Figure 2

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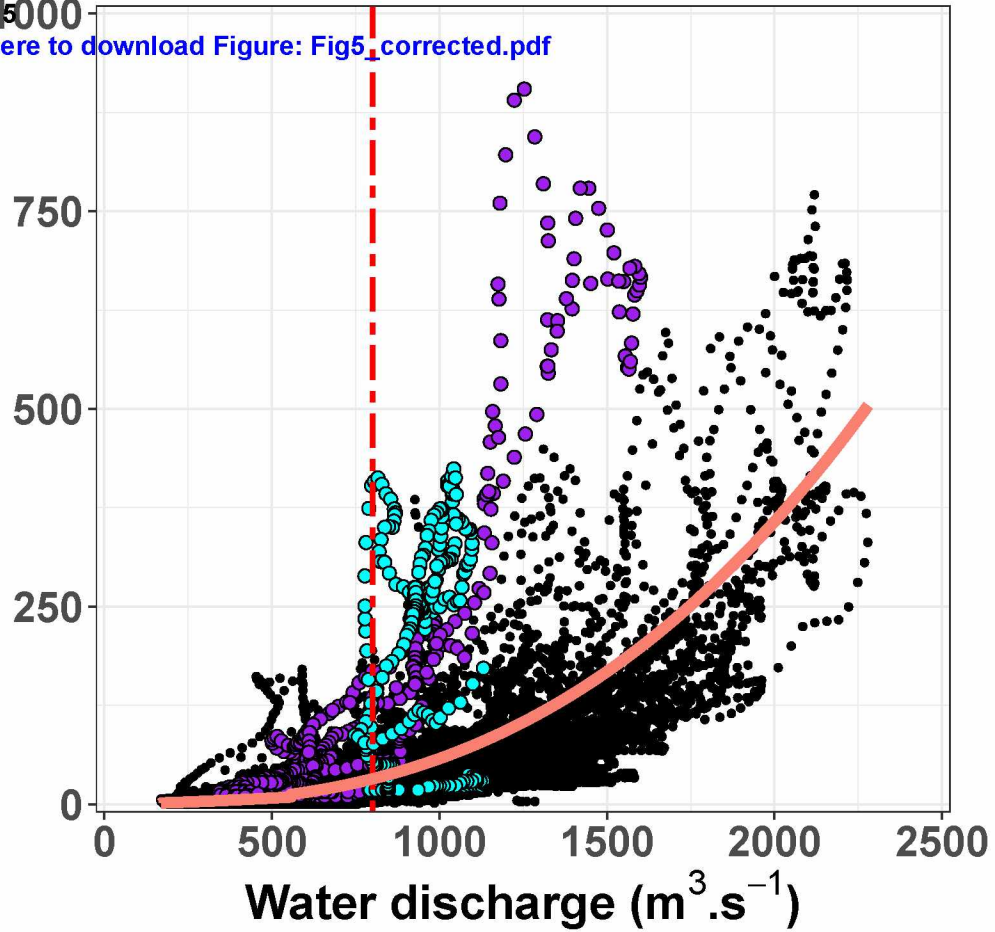
Water level (m)







SPM concentration (mg.L<sup>-1</sup>)



- Flushing operations:
- June 2012
  - May 2016
- ┆ Flood threshold
- Poulier et al. (2019)

Figure 6

[Click here to download Figure: Fig6.pdf](#)

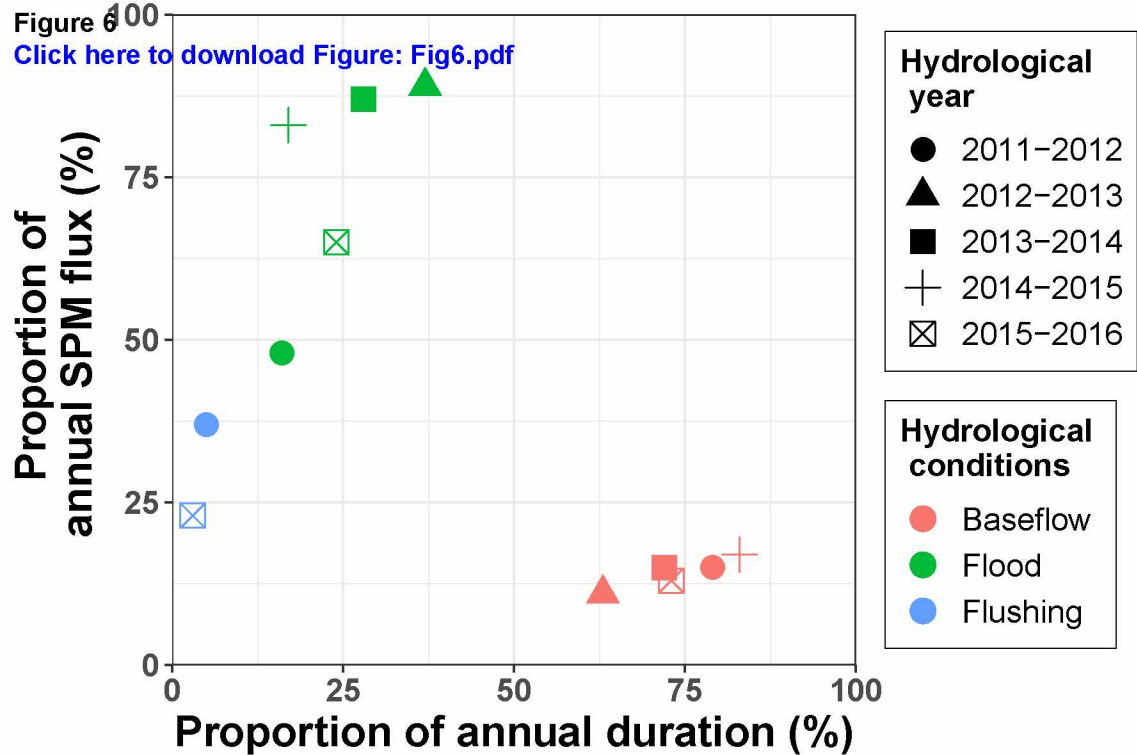


Figure 7

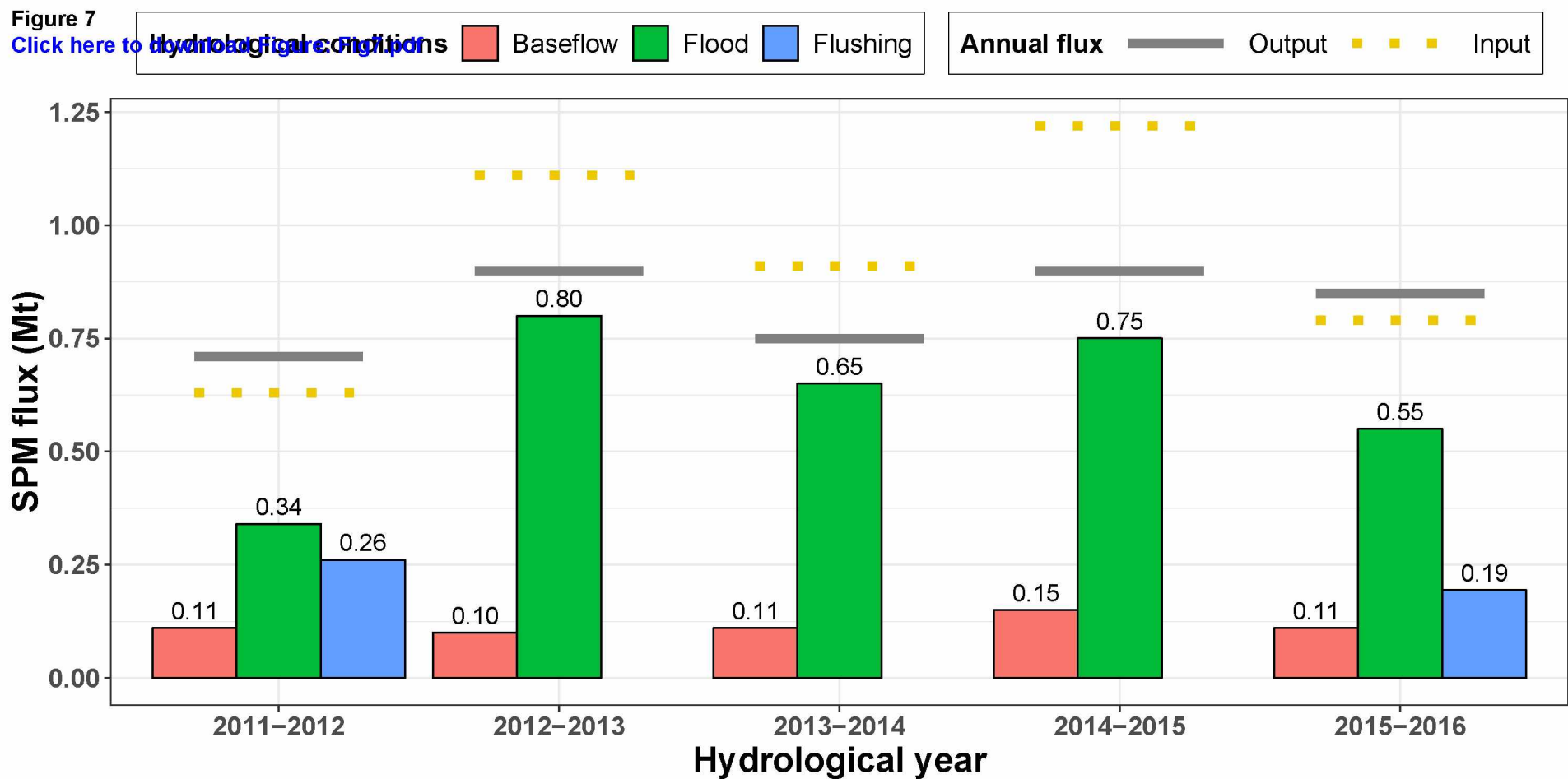
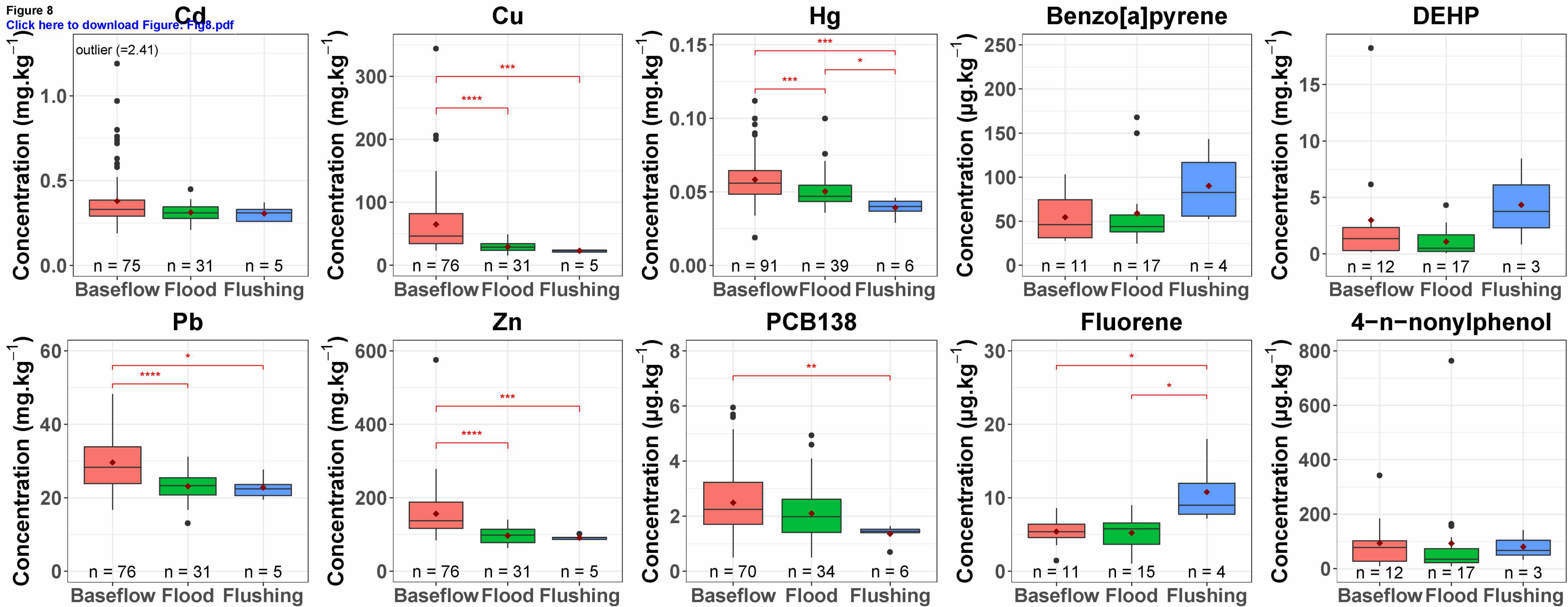
[Click here to Hydrological conditions](#)

Figure 8  
[Click here to download Figure\\_Fig8.pdf](#)





**Table**[Click here to download Table: 7\\_Tables.docx](#)

Table 1 – Proportions (%) of annual SPM and particulate contaminant fluxes in the Rhône River at Jons for the different hydrological conditions (baseflow, flood, flushing event) from September 2011 to August 2012.

Hydrological condition	SPM	Cd	Cu	Pb	Zn	Hg	PCB138
Baseflow	15.5	15.7	15.1	17.1	16.6	14.5	17.5
Flood	47.9	48.9	53.1	49.6	50.7	52.7	55.8
2012 Flushing	36.6	35.4	31.8	33.3	32.8	32.8	26.7

**Supplementary materials**

[Click here to download e-component: 8\\_Sup-Mat\\_corrected\\_v3.docx](#)

**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

## \*Author Contributions Section

Hugo Lepage: Conceptualization, Methodology, Software, Validation, Data Curation, Writing - Original Draft, Visualization

Marina Launay: Conceptualization, Methodology, Software, Formal analysis, Investigation, Data Curation, Writing - Original Draft

Jérôme Le Coz: Conceptualization, Methodology, Resources, Writing - Original Draft

Hélène Angot: Conceptualization, Methodology, Investigation, Writing - Review & Editing

Cécile Miège: Conceptualization, Methodology, Writing - Review & Editing

Stéphanie Gairoard: Conceptualization, Methodology

Olivier Radakovitch: Conceptualization, Methodology, Resources, Writing - Review & Editing

Marina Coquery: Conceptualization, Methodology, Writing - Original Draft, Supervision